A NUCLEAR EMULSION STUDY OF THE α-DISINTEGRATION OF THORIUM AND RADIOTHORIUM, AND OF THE POSSIBLE α-DISINTEGRATION OF MESOTHORIUM 1.

Thesis

submitted by

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Preface.

The research described in this thesis was carried out in the Department of Natural Philosophy of the University of Edinburgh under the joint direction of Professor N. Feather, F.R.S., and Dr. M.A.S. Ross. The results given in Chapter IV have been published, in conjunction with Dr. Ross, in The Proceedings of the Physical Society, A, 1955, 68, 923. A paper based on the results presented in Chapter VI, and prepared in collaboration with Professor Feather and Dr. N. Miller, has been accepted for publication in the same journal. It is intended to submit for publication a paper embodying the results of Chapter V.

Chapter I.

INTRODUCTION AND HISTORICAL REVIEW.

I.1. Introduction.

Meitner and Hahn discovered that certain α-ray bodies emitted homogeneous groups of electrons. Careful experiments showed that these electrons could not have originated in some subsequent β-ray transformation and therefore must have come from the α-ray body. This proved that some process existed by which electrons of definite energies could be liberated from the outer electronic structure during radioactive disintegration. Black (1924) suggested that this process, internal conversion, occurred after disintegration and Meitner (1925) came to the same conclusion.

The results obtained from the studies of the conversion electrons emitted following the radioactive disintegrations of ^{232}Th and ^{228}Th, combined with the available data on the respective α- and γ-radiations, yield information about the properties of the levels excited in ^{228}Ra and ^{224}Ra and illustrate some of the characteristics of α-disintegration in the heavy even-even nuclei.

A schematic representation of the thorium chain in shown in Figure 1.

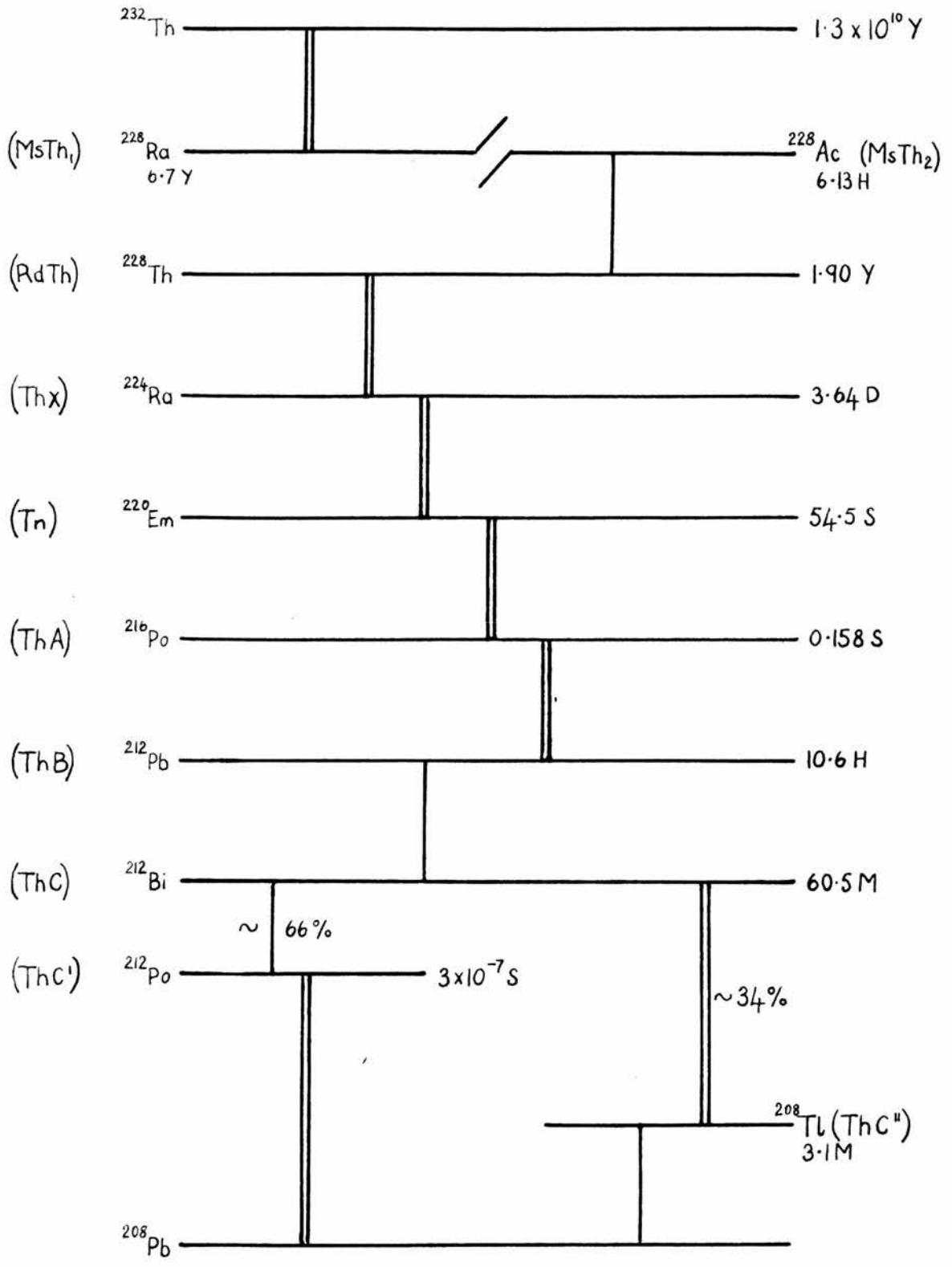

FIGURE 1. SCHEMATIC REPRESENTATION OF THE THORIUM SERIES.

From the data tabulated by Hollander, Perlman and Seaborg (1953).

A single vertical line represents disintegration by β-emission; a double line represents disintegration by α-emission. In the cases in which branching occurs, the fraction following each mode is indicated as a percentage. The letters S, M, H, D, Y stand for seconds, minutes, hours, days and years respectively.

I.2. Historical Review.

I.2.1. ^{232}Th (Thorium).

The α-radiation.

Due to the very long half-life of ^{232}Th, (1.39 $\pm$ 0.05) x 10^{10} years, quoted by Feather 1943 , with the resulting weak specific activity, the accurate determination of the α-particle ranges and therefore of the energies is very difficult. The two lines, which are now known to exist in the α-particle spectrum, have not been resolved experimentally.

In the period between 1911 and 1937 various determinations of the α-particle range were made without disclosing any real agreement. The values quoted for the range in air, at a temperature of 15°C and a pressure of 760 mm. of mercury, lay between 2.53 cm. and 2.90 cm. (see references under Geiger). Feather (1949) quoted the energy determination of Clark, Spencer-Palmer and Woodward (3.976 $\pm$ 0.020 MeV) and the C2 emulsion range of Green and Livesey (15.0 $\pm$ 0.2 μ) and remarked that, since they gave a point which lay clearly on the emulsion range-energy curve, they may be considered as mutually confirmatory. The concentrated half-tone emulsion range of 14.2 $\pm$ 0.2μ, given by Faraggi (1946), supported this conclusion. In 1951 Mlle. Faraggi, using the emulsion range of 14.2 $\pm$ 0.2μ, gave the associated energy as

3.98 ± 0.04 MeV, in good agreement with the above value.

The conversion electron spectrum.

Mlle. Albouy (1952), using Ilford G5 nuclear emulsions impregnated with a solution of ^{232}Th freed from ^{228}Th, studied the conversion electron spectrum. 3,760 α-particle tracks were measured and, from a consideration of their ranges, all were attributed to the α-disintegration of ^{232}Th. Of these events 780 showed an associated conversion electron track, an electron intensity of about 20%. The distribution of the electron ranges showed the existence of two groups, the mean ranges of which were 17 μ and 27 μ, corresponding to energies of 55 keV and 70 keV. Since this energy difference (15 keV) was the same as that between the binding energies of electrons in the L and M shells of ^{228}Ra, Albouy interpreted the observed electron groups as having been emitted from these shells. This led to an excited level of ^{228}Ra approximately 75 keV above the ground state. The mean range of the α-particles with no associated electron was slightly longer than that of those with associated electrons.

Later in the same year Dunlavey and Seaborg (1952) published the results of their investigation of the conversion electrons accompanying the

α-disintegration of some heavy nuclei. From 5,100 ^{232}Th α-particle tracks observed in G5 emulsion, an electron intensity of 24 $\pm$ 3% was estimated. Two groups of electrons were observed with energies of 35 $\pm$ 5 keV and 50 $\pm$ 5 keV. Interpreting them in the same way as above, these authors gave the energy of the excited level of ^{228}Ra as 55 $\pm$ 5 keV. Both Albouy and Dunlavey decided that since the intensity of the γ-ray emission from this excited level was probably very low, the conversion electron intensity was a good approximation to the intensity of excitation of the level.

Philbert, Gènin and Vigneron (1954), from a study of 603 ^{232}Th α-particle tracks in C2 emulsion, gave the mean energy of 3.990 $\pm$ 0.020 MeV, in good agreement with earlier values. On the basis of Albouy's results they estimated that the α-particle spectrum consisted of two lines of energies 4.008 $\pm$ 0.020 MeV; and 3.933 $\pm$ 0.020 MeV. The corresponding energies deduced from the results of Dunlavey and Seaborg were given as 4.004 $\pm$ 0.020 MeV and 3.949 $\pm$ 0.020 MeV.

It was because of the discrepancies between the energies and intensities, assigned to the first excited level of ^{228}Ra by Albouy and by Dunlavey and Seaborg, that the investigation was undertaken by the present author. The results of this investigation

(59 ± 1 keV and 24 ± 3% intensity) were published in 1955.

In a paper of 1956 Albouy revised the values, which she had given previously (1952), to 60 keV and 22 ± 2% intensity.

I.2.2. ^{228}Th (Radiothorium).

The α-radiation.

Estimations of the range and the total effective energy of the ^{228}Th α-particles, as a single unresolved group, have been made by various experimenters. Geiger (1922), revising the earlier measurement of 1911, gave the α-particle range in air, at a temperature of 15°C and a pressure of 760 mm. of mercury, as 4.019 cm. and the energy as 5.347 MeV. Curie (1932), in an experiment with a Wilson cloud chamber, determined the range in air, at the same temperature and pressure, as 3.99 cm. with a probable experimental error of not more than 0.03 cm. Feather (1949) quoted the results of Clark, Spencer-Palmer and Woodward who gave the effective energy as 5.38 ± 0.02 MeV and of Green and Livesey who measured the C2 emulsion range as 22.9 ± 0.2μ. The G5 emulsion range was determined by Albouy and Teillac (1950) as 22.9 ± 0.1μ.

The first workers to observe the fine structure in the α-particle spectrum were Rosenblum and Chamié (1932). Focusing the α-particles with a large

electro-magnet, they found three lines α_1, α_2 and α_x. The two higher energy lines, α_1 and α_2, were definitely attributed to ^{228}Th, but the origin of α_x, a very weak line, was uncertain. The energy differences between the lines were given as $\alpha_1 - \alpha_2 = 82$ keV, $\alpha_1 - \alpha_x = 123$ keV and $\alpha_2 - \alpha_x = 42$ keV. In 1933 these investigators confirmed the existence of α_1 and α_2. The estimated energies of the lines were 5.420 MeV and 5.335 MeV respectively, giving an energy difference, corrected for recoil, of about 86 keV. They suggested that α_x probably originated from Po, which had been removed from the source used. In both these sets of experiments the α-line of ^{212}Bi was used as a standard. Chang and Coor (1948), also using a magnetic analysis method, gave the energy of α_2 as 5.354 ± 0.02 MeV, by taking α_1 (5.430 MeV) as the reference line. Some inconclusive evidence for the existence of weak lines of lower energies was found. The next attempt to analyse the α-particle spectrum was made by Rosenblum, Valadares and Perey (1949). Assuming the highest energy α-line of ^{224}Ra to be 5.681 MeV, they found $\alpha_1 = 5.423$ MeV and $\alpha_2 = 5.338$ MeV, resulting in a nuclear energy difference of 86.7 keV. The intensity ratio of α_2/α_1 was given as $(39 \pm 1)/100$. A slight broadening of the α_1-line was observed.

The results of a detailed study of the complex α-particle spectra of the heavy elements have been published by Asaro in a thesis of 1953. The spectrum of the ^{228}Th α-particles was given in a paper by Asaro, Stephens and Perlman (1953). The α-particle spectrograph employed a 60° symmetrical magnetic analyser, the normal trajectory having a radius of curvature of 75 cm. The source used was purified by the use of a Dorvex-50 ion exchange resin, upon which the radium and thorium fractions were adsorbed and eluted selectively. After the chemical separation of a sample, its solution was evaporated on a tungsten filament, and then vacuum sublimed on to a platinum plate marked to approximate to a line source of α-activity. The α-particles were detected in nuclear emulsion plates. After exposure the plates were examined under a microscope and a count made of the number of tracks whose direction lay within a certain cone of acceptance. Taking the α-line of ^{224}Ra (5.681 MeV) as a standard, the four lines observed were associated with energies of 5.421 ± 0.001 MeV (α_1), 5.338 ± 0.001 MeV (α_2), 5.208 MeV (α_3) and 5.173 MeV (α_4), which gave nuclear energy levels at 84.3 keV, 217 keV and 253 keV above the ground state. The respective intensities were estimated as 71%, 28%, 0.4% and 0.2%. The energy and intensity values given

for the α_1 and α_2 lines agreed well with those found by Rosenblum, Valadares and Perey.

The γ - and X-radiations.

Thibaud (1926), by a rotating crystal method, found two electromagnetic radiation lines with wavelengths of 1.45×10^{-9} cm. and 1.68×10^{-9} cm. Meitner (1929), on the basis of the results obtained from a study of the conversion electron spectrum, concluded that the 1.68×10^{-9} cm. line could not be attributed to the disintegration of ^{228}Th and by repeating the above experiment showed that it originated from ^{212}Pb.

Riou (1949 and 1950), using a selective absorption technique with a Geiger-Müller counter, filled with xenon and alcohol, as a detector, studied the electromagnetic radiation spectrum emitted from a pure source of ^{228}Th. Three lines were found with energies and intensities (number of photons per 100 disintegrations) of 14 keV and 7 ± 1.5, 83.3 keV and 1.8 ± 0.5 and 86.8 keV and 0.7 ± 0.2. By considering the difference in binding energies of electrons in the L-and M-shells of ^{224}Ra, the 14 keV line was shown to be the L X-ray of radium. It is also emitted following the disintegration of ^{230}Th. From the intensity ratio of $\frac{\text{86.8 keV line}}{\text{83.3 keV line}}$ (≈ 0.35), Riou remarked that these lines could not be the K X-rays of

radium, since in normal emission $\frac{\text{Intensity of } K\alpha_1}{\text{Intensity of } K\alpha_2} = 2$. He therefore interpreted them as two γ-rays emitted from an excited level of ^{224}Ra at about 87 keV above the ground state. This interpretation fitted in with the apparent broadening of the α_1-line observed by Rosenblum, Valadares and Perey, which suggested the possibility of the existence of a level very close to the ground state, as shown in Figure 2. Using the intensity of the L X-ray and the fluorescent yield (0.37), Riou calculated that the L-shell should be ionised in (19 $\pm$ 3)% of the disintegrations, a result in violent disagreement with those of Surugue and Tsien, but in good agreement with the results of later work.

In 1951 Beling, Feld and Halpern studied the α- γ angular correlation. The α-particles and the γ-rays were detected by scintillation counters and differential discrimination was used on both α- and γ-pulses. A large number of runs, each lasting several hours and including angles in all four quadrants, were made on thin sources of ^{228}Th freshly separated from a mixture of ^{228}Th and its disintegration products. They supposed, in accordance with current ideas, that two γ-rays were emitted. Absorption measurements of the coincidences showed that the γ-rays had energies between the K-edges of gold and

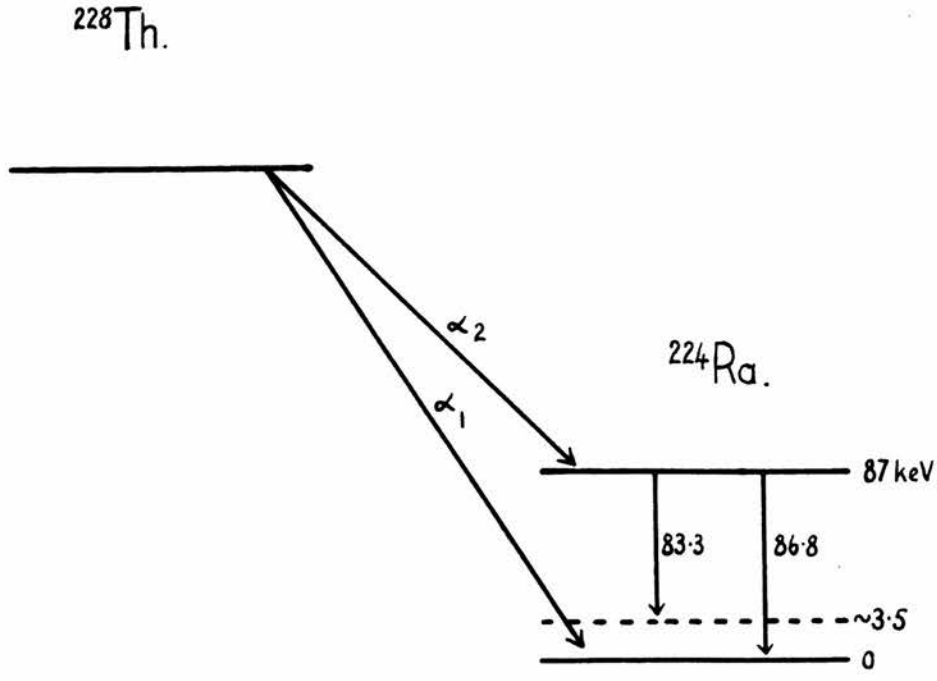

FIGURE 2.

lead (81 keV and 88.5 keV respectively) and that their intensity relative to the α-particle intensity was roughly as expected. The fact that coincidences were observed with a 0.3 μ sec. resolving time and the large $\cos^4 \theta$ (θ = angle between the detectors) term in the observed correlation, suggested that the radiation was E2. By absorbing the 86.6 keV radiation in thallium separate correlations were obtained for the two radiation lines. The results showed fairly good agreement with the theoretical correlation, and although the differences were significantly outside the experimental errors, these workers concluded that the spin sequence $0 \xrightarrow{\alpha} 2 \xrightarrow{\gamma} 0$ at least played an important part in disintegration of $^{228}Th \longrightarrow {}^{224}Ra$. Battey, Madansky and Rasetti (1953), supposing only one γ-ray (after Riou 1953), also studied the α - γ angular correlation and arrived at a similar conclusion. In the same year Abragam and Pound (1953) published a paper in which they suggested that the discrepancies between the theoretical and observed correlations could be explained by the quadruple coupling in the intermediate state.

The first conclusive evidence of the existence of higher levels in ^{224}Ra which are excited by the α-disintegration of ^{228}Th was given in a paper by Bouissières, Falk-Vairant, Riou, Teillac and Victor

(1953). Detecting the γ-rays from a pure source of ^{228}Th with a sodium iodide, thallium activated crystal, they found four γ-lines in the spectrum. Using the 283 keV ray of ^{203}Hg and the 140 keV ray of $^{99}Tc^{x}$ as reference lines, the energies were given as 84 keV, 133 $\pm$ 2 keV, 172 $\pm$ 4 keV and 216 $\pm$ 3 keV, with the relative intensities of 100, 14, 10 and 17 respectively. In a communication to those workers Rosenblum mentioned that in the α-particle spectrum a line probably existed which would lead to an excited level of 216 keV. They interpreted the 133 keV γ-ray as belonging to the transition from the 216 keV level to the 84 keV level. The relative intensities of the 133 keV γ-ray and the 216 keV γ-ray led them to suggest that the 216 keV level was a 2 + state. This was suggested also by Falk-Vairant in a thesis of 1954. These experiments also observed the L X-ray of ^{224}Ra at 15 keV.

Riou (thesis 1953) gave experimental results which differed considerably from those of his earlier work. Only two radiation lines were quoted as belonging to the ^{228}Th disintegration, the 15 keV L X-ray and one γ-ray of 85 keV. The percentage intensities were given as 7 $\pm$ 1.5 and 2.1 $\pm$ 0.7 respectively. From the γ-ray intensity and the available information from the α-particle and conversion electron spectra, he calculated that the total intensity of the conversion

electron emission should be about 25%, which led to a value of 18 ± 1% for the intensity of the emitted L-shell electrons. This value was in good agreement with that of 17 ± 4% deduced from the L X-ray intensity and the fluorescent yield 0.40, previously given as 19 ± 3% (1949 and 1950). Using the intensity of 18 ± 1% Riou calculated the internal conversion coefficient for the L-shell as 10 ± 4, and from a comparison with the theoretical coefficients he considered that the radiation was E2. The spectroscopic designation (2 +) of the 85 keV level of ^{224}Ra therefore conformed to the empirical rules regarding the first excited levels of the heavy even-even nuclei.

With a freshly separated source of ^{228}Th (within 1 hour of use) Asaro, Stephens and Perlman (1953) studied the γ-ray spectrum by using a sodium iodide, thallium activated crystal as a detector. The purified solution was evaporated down, alpha counted and pulse analysed in a 50-channel gamma-scintillation counter. The α-particle activity of the source used was 3×10^6 disintegrations per minute. Four lines were observed with energies of 89 keV, 137 keV, 169 keV and 212 keV. These energies were estimated by using the 60 keV γ-ray of ^{241}Am and the 184 keV γ-ray of ^{235}U as reference lines and were probably accurate to about 5 keV. By assuming that one 60 keV ^{241}Am

γ-ray was associated with 40% of the ^{241}Am α-disintegrations, the respective abundances of these γ-rays, after relevant corrections had been made, were found to be 1.6, 0.26, 0.09 and 0.27% of the ^{228}Th α-emission. The 89 keV γ-ray was attributed to the de-excitation of the 84 keV level, and since this level was populated by an α-particle feed of 28% intensity, an internal conversion coefficient of 16 was calculated. This value strongly suggested that the radiation was E2 and therefore the spectroscopic designation of 2 + was given to the first excited level of ^{224}Ra. The 169 keV γ-ray corresponded to a transition from the 253 keV level to the 84 keV level. From a consideration of the intensity of excitation of this higher level (0.2%) the internal conversion coefficient was found to be 1.2. This led these workers to the conclusion that the radiation was E2 and they therefore assigned a spin of 0, 2 or 4 and even parity to the 253 keV level. Because of the absence of a cross-over transition and the apparent absence of any M1 admixture, the designation of 4 + was thought to be the most probable.

The energies of the 137 keV γ-ray and the 212 keV γ-ray agreed, within the experimental errors, with the transitions from the 217 keV level to the 84 keV level and to the ground state respectively. The sum

of the intensities was the same (greater from the experimental results) as the population (0.4%) of this 217 keV level and so the conversion coefficients for both γ-rays must be < 1. This could be fulfilled only by E1 radiation and therefore it was suggested that the 217 keV level was a 1-state.

The intensities quoted by Asaro (thesis 1953) and by Stephens (thesis 1955), although differing slightly from the above, were substantially the same.

Additional evidence for the conclusion that the second excited level of ^{224}Ra is a 1-state was provided by the α- γ angular correlation. The results of the investigation of the observed correlation of the 212 keV γ-rays with the α-particles feeding the 217 keV level were given by Stephens, Asaro and Perlman (1954) and in a thesis by Stephens (1955). The observed correlation did not agree exactly with the theoretical correlation but the discrepancy probably was due to the large sample size and to the relatively poor geometry, for which no corrections were made. Nevertheless, the experimental results were taken as fairly good support for the assignment of 1 - to the second excited level, particularly as there were limitations on other possible assignments. It was fairly certain that the 212 keV γ-ray went to the ground state of ^{224}Ra and therefore must be electric. The assignment of E3 or

higher multipolarities was ruled out on the basis of the prompt coincidences observed. These workers, therefore, considered only E1 or E2 and the angular correlation expected for E2 is very different from that found. The γ-radiation from the 84 keV level was confirmed as being probably E2.

Early in 1954 Newton and Rose gave the results of their investigations on the γ-ray spectrum between 30 keV and 220 keV. The energies and relative intensities of the 4 lines, which they attributed to the de-excitation of the ^{224}Ra nuclei, were given as 84.4 ± 0.2 keV (1), 132.3 ± 1.3 keV (0.105 ± 0.010), 167 ± 2 keV (0.058 ± 0.012) and 214 ± 3 keV (0.18 ± 0.05), in good agreement with previous results. The level scheme and the spectroscopic designations which they favoured lent support to the conclusions of Asaro, Stephens and Perlman, although they suggested that the third excited level might lie 166 keV above the 217 keV level and have an energy of 383 keV.

The conversion electron spectrum.

Using a magnetic spectrograph, Meitner (1929) studied the conversion electron spectrum emitted following the disintegration of ^{228}Th and found four intense lines and two very weak lines. She interpreted the four strong lines as having been produced by two radiation lines, the energies of which were the same as

those of the Kα_1 and Kα_2 X-rays of ^{224}Ra. Thus, Meitner supposed that the ^{228}Th α-particles, in passing through the electron shells, excited the K-radiation, which in turn caused the emission of Auger electrons from the L- and M-shells. A difficulty which arose from this interpretation was discussed by Riou (1949).

In 1941 Surugue and Tsien found four conversion lines, the energies of which agreed fairly well with those of the strong lines observed by Meitner. The existence of the two weak lines was not confirmed. At this time two lines were known to exist in the α-particle spectrum and so these workers postulated the existence of two γ-rays of energies 83.3 keV and 86.8 keV, the electrons having been emitted from the L- and M-shells. The total intensity of the conversion electrons was given as 6.9% of ^{228}Th α-emission.

A nuclear emulsion study of the conversion electrons was made by Albouy and Teillac (1950). Impregnating Ilford G5 electron sensitive emulsions with solutions of pure ^{232}Th and ^{228}Th in equilibrium with its disintegration products, they identified 1850 events as belonging to the α-disintegration of ^{228}Th, of which 141 showed associated conversion electron tracks. The energy of the electrons emitted from the L-shell was given as 65 keV which led to an excited

level of about 83 keV. The electron intensity, of approximately 7.6%, agreed well with that observed by Surugue and Tsien, but when considered in conjunction with the γ-ray intensity of Riou (1949) showed no agreement with the results obtained from the α-particle spectrum.

Rosenblum, Valadares and Guillot (1952), by means of a β-spectrograph, re-examined the conversion electron spectrum. The electrons were detected in plates which, after exposure, were scanned with a microphotometer. Four lines were observed, two of which were shown to be doublets, but the two weak lines found by Meitner were not seen. The results are given in Table 1.

Table 1.

	H (oe.-cm.)	Electron energy (keV)	Relative intensity	Ionisation energy (keV)	Energy of the excited level (keV)
A	892	65.8	100	18.5 (L_{II})	84.3
B	914	68.9	84	15.4 (L_{III})	84.3
C	988	79.7	29	4.5 (M_{II})	84.2
C_1	993	80.4	23	3.8 (M_{III})	84.2
D	1012	83.3	13	1.1 (N_{II})	84.4
D_1	1018	84.0	2	0.3 (0)	84.3

F ray of ^{212}Pb 1388.5 oe.-cm. used as reference line.

The lines A, B, C and D corresponded to those found by Meitner and by Surugue and Tsien. Rosenblum, Valadares and Guillot pointed out that the previous interpretations which presumed the existence of two radiation lines, either X-rays or γ-rays, necessitated that the energy differences B - A and D - C should be the same, whereas their results showed that the former energy difference was 0.5 keV less than the latter. From the ratio of the number of electrons emitted from the L_{II} and L_{III} sub-shells they deduced that the observed spectrum could be interpreted on the basis of only one γ-ray of energy 84.3 keV and multipolarity E2. The sub-shells M_{II}, M_{III}, N_{II} and O were chosen to fit the energy given by the conversion in the L_{II} and L_{III} sub-shells. They therefore concluded that ^{224}Ra had only one low-lying excited level, 84.3 keV above the ground state, with a spin 2 and even parity. In 1954 they repeated this investigation and found substantially the same results, although the energy of the excited level was revised to 84.5 keV.

With a coincidence counting technique Victor, Teillac, Falk-Vairant and Bouissières (1952) determined the number of conversion electrons emitted per α-disintegration. Using the photon intensity given by Riou the total internal conversion coefficient α_T was found to be 12 compared with the theoretical

coefficient for E2 of 16 (Gellman, Griffiths and Stanley). They therefore considered that the radiation was E2. Falk-Vairant (thesis 1954) carried this calculation one step further and arrived at the same conclusion. From the results of Rosenblum, Valadares and Guillot, which showed that 74% of the electrons were emitted from the L-shell, he found $\alpha_L = 8.9 \pm 5$.

Jarvis (1953), using Kodak NT4 nuclear emulsions, impregnated with ^{228}Th solution, found electrons which appeared to be emitted in the de-excitation of the higher energy levels of ^{224}Ra. She considered 569 α-particle tracks of which 214 showed associated conversion electrons, an intensity of 42%. The two highest energy lines (27% intensity) represented electrons emitted from the L- and M-shells and indicated an excited level at about 84 keV. These electrons were associated with α-particle tracks with a mean range 0.5 μ shorter than the normal, which corresponded to an energy difference of 87 ± 24 keV. This intensity, along with the γ-ray intensity of Riou, gave a total intensity of excitation of the 84 keV level in good agreement with that obtained from the α-particle spectrum. A group of low energy electrons, with an intensity of 15%, were associated with α-particles of a mean range 1.2 μ less than the normal and this suggested that these electrons were emitted in the

de-excitation of a level 209 ± 28 keV above the ground state. Jarvis concluded that this was consistent with higher energy γ-rays found by Bouissières and his collaborators (1953), since the electrons could be attributed either to the L-conversion of a 44 keV γ-ray (216 keV - 172 keV) or the K-conversion of the 133 keV γ-ray. The intensity, however, was very much higher than allowed by the α-particle feeds to these levels. A few events which indicated a cascade process were observed but not discussed.

Albouy (1956), from a nuclear emulsion study, gave the energy and intensity of the first excited level as 84 keV and 25 ± 2% and concluded that the radiation was E2.

Summary.

The results of these investigations, on the whole, are consistent with the level scheme suggested by Asaro, Stephens and Perlman (see Figure 3), although the quoted energies, with the exception of that of the first excited level, may be subject to slight corrections. The implications of this scheme will be discussed in Chapter V. The first excited state is almost definitely a 2 + state. Two spectroscopic designations, 2 + and 1 -, have been suggested for the 217 keV level but the weight of evidence favoured 1 -. On the assumption that the third excited level was

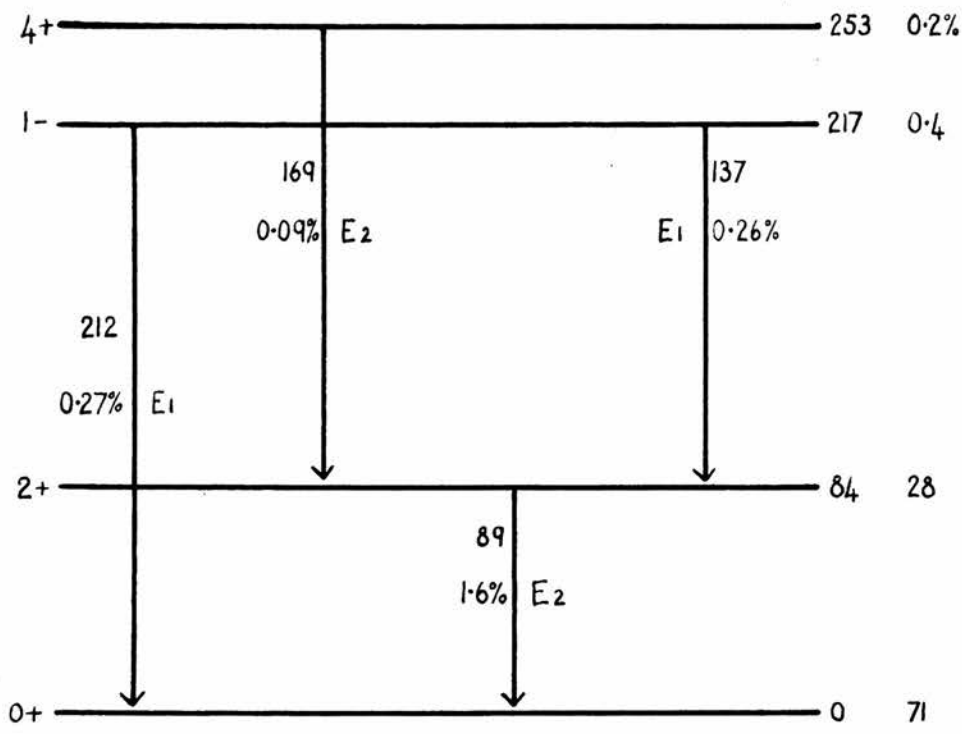

FIGURE 3. THE ^{224}Ra LEVEL SCHEME SUGGESTED BY ASARO, STEPHENS AND PERLMAN (1953).

de-excited only by a transition to the 84 keV level, the designation of 4 + was thought to be the most probable.

This was the position when the present investigation was started.

I.3. Outline of the present work.

Ilford G5 electron sensitive emulsions (200 μ thick) were impregnated with the active solutions, exposed, processed and then examined under a microscope. In both investigations, the events considered consisted of a single α-particle track, the α-particle "stars" which resulted from successive disintegrations in the thorium chain being ignored. Following the α-disintegration of ^{232}Th and ^{228}Th, approximately 25% of the daughter nuclei are left in an excited state. The excited levels are de-excited either by the emission of γ-rays or by the emission of conversion electrons. These electrons can be recorded by the nuclear emulsion and thus events were observed in which an α-particle track and an electron track had a common origin. Since the nuclear emulsion technique permitted a study of the disintegrations one at a time, information regarding the energies and absolute intensities of the conversion electron lines was obtained.

Chapter II.

EXPERIMENTAL METHOD.

II.1. Preparation of Sources.

To reduce the number of unwanted events in the emulsion, the sources used were prepared as free from contamination as possible.

Chemical Separations.

^{232}Th. To a few drops of a solution of ^{232}Th with its decay products (not in equilibrium) 2 mg. each of aluminium and barium carriers were added. The aluminium hydroxide was precipitated with NH_4OH, carrying with it almost all the thorium and leaving almost all the radium in solution. The precipitate was centrifuged down, washed three times to remove traces of radium, and dissolved in 5 ml. of 1N HCl. 2 mg. of bismuth carrier were added and precipitated as bismuth sulphide by bubbling H_2S through the solution to saturation. This removed ^{212}Pb (ThB) and ^{212}Bi (ThC) as sulphides. The precipitate was filtered off and boiled gently for about 5 minutes to remove the H_2S. 2 mg. of barium were again added and the aluminium hydroxide precipitated with ammonia. The precipitate was then centrifuged down, and redissolved in 2 - 3 drops of concentrated HCl, the solution being

subsequently made up to 5 ml. with distilled water. Finally, the precipitate of hydroxide was washed four times to remove HCl and dissolved in a few ml. of a 10% solution of citric acid.

^{228}Th. Two techniques were used in this case. For the first impregnation (Plate 1) the above method was used starting with a solution of ^{228}Th in equilibrium with its decay products. Difficulty arose in the investigation of some events due to the apparent presence of single ^{224}Ra α-particle tracks. This point will be discussed more fully in Chapter V. In an attempt to effect the complete removal of ^{224}Ra by recycling the latter stages of the separation four times, it was found that the ^{228}Th, due to the small mass involved, was lost in the process. To overcome this difficulty the following method was used for the second impregnation (Plate 2).

To a few drops of the ^{228}Th solution 1 mg. each of barium and lead "hold-back" carriers and 3 mg. of cerium carrier were added. The ceric carrier was precipitated as iodate by iodic acid in 0.1 N HCl solution. This precipitate, which carried the ^{228}Th, was centrifuged down, washed four times and dissolved in 2 ml. of 1N HNO_3. Following this, the cerium was reprecipitated as hydroxide with NH_4OH. Subsequently the hydroxide precipitate, which carried the ^{228}Th, was

dissolved in 5 ml. of dilute HCl and reprecipitated with NH_4OH, this process being recycled a further two times to remove traces of HIO_3. Finally the precipitate was washed four times and dissolved in a few ml. of a 10% solution of citric acid.

All the impregnations were done immediately following the separations, using emulsions which had been made a few days previously.

II.2. Treatment of Plates.

II.2.1. Impregnation of Plates.

The reason for the presence of the citrate ion in the impregnating solution is twofold. It has been found (Yagoda, 1949; Jarvis, 1950) that the impregnation of nuclear emulsions with heavy elements is complicated by the tendency of these elements to form chemical bonds with the outer layers of the emulsion. The citrate ion forms a complex with the heavy element which prevents this bonding, and so permits effective penetration of the impregnating material (Zajac and Miller, 1952). The citrate also prevents the precipitation of barium in the emulsion when carbonates are added during development.

Ilford G5 electron sensitive emulsions yield maximum sensitivity when impregnated with a neutral solution, and therefore the solution of the active

material should have pH ~ 7 when possible. In the case of ^{228}Th this value of the pH was used. With ^{232}Th, however, the longer half-life necessitated the use of a larger mass of active material and this led to complications. Due to the variation of the residual solubility of thorium citrate with the pH of the aqueous solvent, an acid solution had to be used to obtain a uniform density of active material throughout the complete depth of the emulsion. A pH between 3 and 4 gave satisfactory results.

Since, in both investigations, the exact activity of the source used was unknown, it was necessary to perform tests covering a wide range of source dilution, in order that a satisfactory density of events be obtained in the final plates. The plates (emulsion thickness 200 μ) were cut into 1 in. x 1 in. sections and soaked in the radioactive solution for 45 minutes at room temperature. The sections were then dried for 2 hours in a current of warm air (about 30°C) and stored for various lengths of time in the presence of a saturated solution of K_2CO_3 (relative humidity 44%).

The soaking of the plates in water before impregnation, in order to swell the emulsion, made no appreciable difference to the speed of penetration of the impregnating material. This stage was dispensed with in the preparation of the plates finally used in

the investigations. As the emulsion dried it tended to peel off from the glass backing. This may have been due to the mechanical stresses set up in the gelatine as it contracted. To prevent the emulsion actually peeling off, great care was taken in the cutting of the plates and the drying was not allowed to be too rapid. The addition of a few per cent. of glycerine to the impregnating solution had no appreciable effect.

II.2.2. Exposure of Plates.

Since the initial energies of the particles studied were deduced from their ranges, it was desirable that all disintegrations should have taken place while the stopping power of the emulsion was constant. The stopping power of the emulsion depends upon its moisture content which, in turn, depends upon the relative humidity of the surrounding air. It was therefore essential to control the relative humidity during the exposure time of the plates. The value of 44% was that normally used in the laboratory.

The choice of exposure times used was influenced mainly by two considerations. The emulsion records the tracks of ionising particles during the impregnation, drying and development times, as well as during the exposure time. The ranges of tracks recorded in these conditions of variable stopping power will

differ materially from those recorded when the emulsion is "normal" (during the exposure time). In order to prevent serious trouble from this source, it is necessary to reduce the ratio of such events to the total number of events as much as possible. This entails the use of a total exposure time which is long compared with the wet (impregnation plus drying plus development) time. The second consideration, the cosmic ray background, unfortunately acts in the opposite direction; the longer the exposure time the greater is this background. Thus, an optimum had to be found. After several trials, an exposure time of 10 days appeared to be a reasonable approximation to this optimum, and was used in both investigations. The consideration of half-lives was not critical in the present work.

II.2.3. Processing of Plates.

The plates were developed according to the temperature development method of Dilworth, Occhialini and Payne (1948). The solutions used are given in Table 2.

The 'Elon' developer used does not give satisfactory results unless the pH of the plates is neutral. This effect has been found by Picciotto (1949) working with uranium loaded plates. In order to neutralise the plates used in the ^{232}Th investigation, they were

soaked, before development, in a sodium bicarbonate solution for 20 minutes. The developer was chilled to approximately 4°C (temperature of water at its maximum density). The plates, still immersed in the sodium bicarbonate solution, were gradually cooled down to this temperature. They were then transferred to the developer and left for 30 minutes. During this time the developer diffused throughout the emulsion but did not react chemically with it to any appreciable extent. The developer plus plates were then transferred to a warm bath (temperature 22°C) and left for 25 minutes while development proceeded. This time, chosen after a series of trial development times had been used, was found to produce plates in which the internal conversion electron tracks were fully developed and yet caused the formation of relatively few fog grains. Next, the plates were put into the stop bath for 20 minutes. At the end of this time the layer of silver present on the surface of the emulsion was removed by gentle rubbing with a moist finger. Finally, the plates were transferred to the fixing bath and left there for one and a half times the length of time required to produce complete clearing. During this time the "hypo" and plates gradually cooled from 22°C to room temperature.

The same procedure was followed with the ^{228}Th

plates with the exception that pure water replaced the sodium bicarbonate soaking solution.

The plates were then washed to remove the hypo. The hypo concentration was gradually reduced by the addition of water at room temperature. This reduction was continued until a sample of the wash water was no more effective in decolourising a dilute $KMnO_4$ solution than was an equal volume of tap water. (For details of this test see Ilford Manual (January 1953) page 211.) After this the plates were laid out horizontally and allowed to dry.

When the plates were dry they were soaked in the glycerine solution for 2 hours. This was an attempt to replace the glycerine lost from the emulsion during processing and so toughen the emulsion, making it less likely to peel off in the dry atmosphere of the laboratory.

Some plates were developed using "Amidol" developer, but the results obtained were not so satisfactory as those obtained with "Elon".

II.3. Microscopic Examination of the Plates.

The plates were studied under a Watson-Bactil binocular microscope (2 mm. oil immersion objective, x 10 holoscopic eyepieces), the rated overall magnification being 980 diameters. Photographs of some

representative events are shown in Figure 4.

As stated in Chapter I, only single α-particle tracks (with or without associated conversion electron tracks) were studied. This limitation was imposed so that the identification of a conversion electron track as being associated with the α-particle track under consideration could be made. This would not be possible if α-particle stars were considered (see Figure 4a). To reduce the possibility of missing a conversion electron track associated with an α-particle track, only those events which originated at least 20μ below the emulsion surface, or 20μ above the glass backing, were studied.

The actual numbers of events studied in the investigations are given in the relevant chapters.

The three-dimensional ranges of all single α-particle tracks were measured, and the initial energies of the α-particles estimated from the mean range versus energy calibration of Rotblat (1950). From the known energies* of the α-particles emitted in the various naturally occurring radioactive disintegration those emitted from ^{232}Th and ^{228}Th were identified. Similar measurements were made on the associated

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* The energies are given in the Table of Isotopes (Hollander, Perlman and Seaborg, 1953).

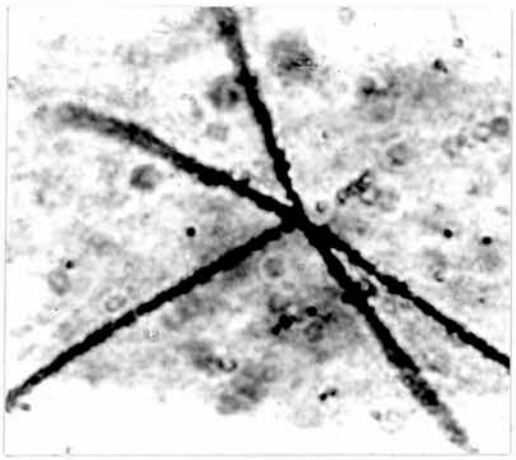

FIGURE 4a. An α-particle "star" formed by successive disintegrations starting at ^{228}Th.

It is impossible to determine with which α-particle track a conversion electron is associated.

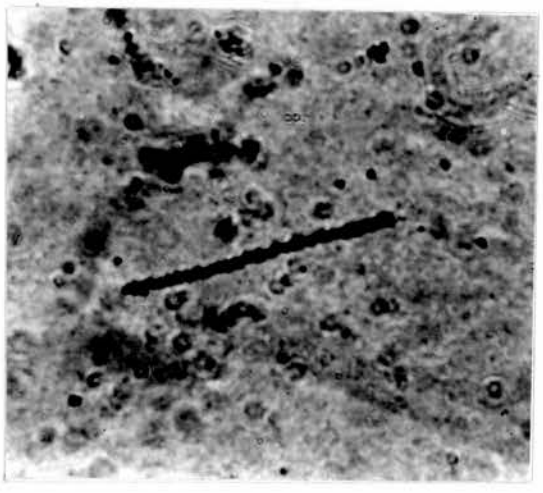

FIGURE 4b. A ^{228}Th α-particle track with no associated electron track.

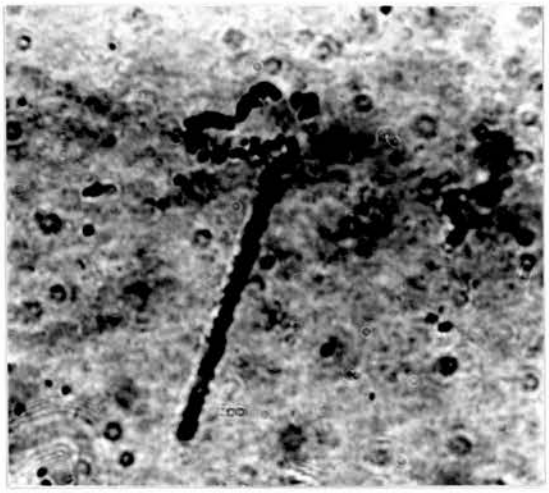

FIGURE 4c. A ^{228}Th α-particle track with associated conversion electron emitted in the de-excitation of the 84 keV level of ^{224}Ra.

Although the events shown above are associated with the disintegration of ^{228}Th, the corresponding events associated with the disintegration of ^{232}Th look very similar, with the slight difference that the α-particle and conversion electron ranges are shorter. ~~and the "star" consists of six α-tracks.~~

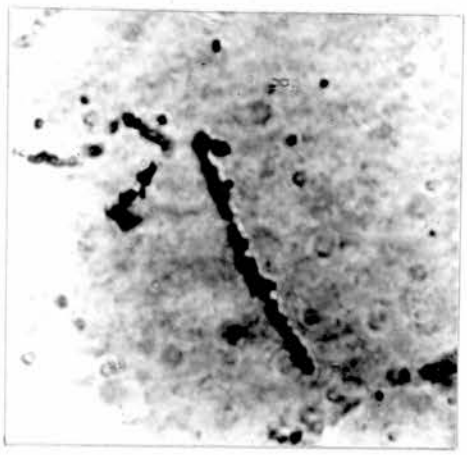

FIGURE 4d. A ^{228}Th α-particle track with an associated conversion electron interpreted as having been emitted in the de-excitation of a higher level (see V.4.5).

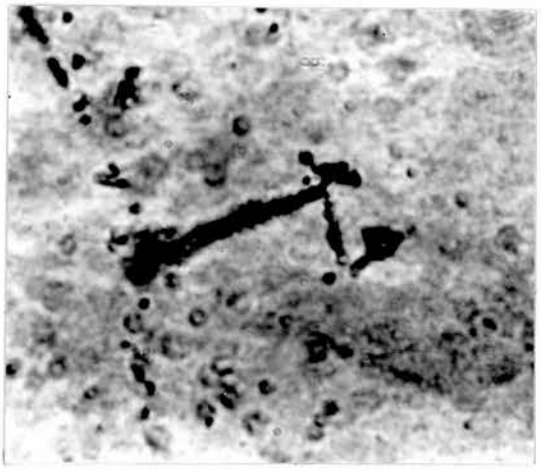

FIGURE 4e. A ^{228}Th α-particle track with two associated conversion electrons indicating a cascade process. The electrons were interpreted as having been emitted in the de-excitation of the 217 keV level (see V.4.2).

conversion electron tracks. Grain counts were also taken for these electron tracks.

In estimating the three-dimensional range of an ionising particle, the track is first analysed into straight line segments, the length of each is obtained from measurements of its horizontal and vertical projections, and these lengths are summed to give the total track length. In the case of an α-particle track, the entire track usually consists of just one segment. The path of an electron is more tortuous, especially near the end of its range, and so the estimate of its length requires the measurement of a large number of segments.

Measurements of the horizontal projections were performed with a calibrated eyepiece graticule (approximately 1 μ per division). The vertical projection measurements involved the use of a graduated fine-focusing adjustment (approximately 0.8 μ per division), which was always racked upwards in order to minimise the mechanical lag. In both cases the scales could be read to the nearest 0.1 μ, but this does not reflect the true accuracy of the measurements.

In accordance with the current practice, the measurements were made between the centres of the grains that defined the ends of the segment. There is no guarantee that the population of track lengths

so obtained coincides exactly with the population of true ranges. The relationship between the two populations is hopelessly complicated, and only the former is obtainable experimentally. Superimposed on this uncertainty are the difficulties of measuring the small distances which are often involved; this is especially true in the depth measurements where the finite depth of focus of the microscope, and the imperfections of the focusing mechanism limit the accuracy.

Chapter III.

MEASUREMENTS ON α-PARTICLE AND INTERNAL CONVERSION ELECTRON TRACKS.

III.1. α-Particle Measurements.

III.1.1. Introduction.

The three-dimensional range of an α-particle track is given by $R = (x^2 + y^2)^{\frac{1}{2}}$, where x is the true horizontal projection of the range and y is the true vertical projection of the range. The problem to be discussed now is that of relating the range projections which are measured in the processed emulsion to those produced in the emulsion during the exposure time.

During the processing of nuclear emulsion shrinkage occurs. This effect has been studied by Powell (1946), who found that no appreciable change occurs in the length or the width of the emulsion, but that the removal of silver bromide during fixation causes a reduction in the emulsion thickness. The amount of this reduction depends on the initial composition of the emulsion.

Thus, while the accurate measurement of x presents no difficulty, the accurate determination of y requires a knowledge of the shrinkage factor, k. It is then necessary to determine k and to study its variation with position in the emulsion and with steepness of

track.

Rotblat and Tai (1949) found that k is independent of depth in the emulsion. They also found that the value of k obtained from the ratio of the thicknesses of the unprocessed and processed emulsion caused the ranges of α-particle tracks to increase with track steepness, whereas it is expected that the true ranges would be independent of steepness. They concluded that this value of k is applicable to tracks whose angle to the horizontal is less than about 30°, but that a smaller value must be used for steeper tracks. Jenny and Hürlimann (1951) showed that k is independent of both depth in the emulsion and steepness of track, and they pointed out that if Rotblat and Tai had used a smaller value of k the observed dependence on steepness would have vanished. Horan (1953), in agreement with the results of Rotblat and Tai, observed an increase in length with steepness, but Greenberg and Haslam (1953) have remarked that if Horan had used a smaller value of k he would have observed none. These latter authors, like Jenny and Hürlimann, reached the conclusion that k is independent of depth and steepness. Work by Palmer and Simons (1955) showed no variation of track length with angle of dip.

The reason suggested by Rotblat and Tai (1949) for the observed dependence of k on steepness is that

during shrinkage the grains on a steep track come into contact with each other and are so prevented from following further displacements of the emulsion. Jarvis (1950), using heavily developed plates, obtained results in agreement with this suggestion. In the present work the plates used were not heavily developed, and it was assumed that k is independent of both depth and steepness. The results obtained indicate that this was justified.

III.1.2. Experimental Method.

The method used in the present investigations to estimate the range of an α-particle track is based on a method, suggested by Vigneron (1949), to determine the ~~shrinkage~~ depth conversion factor of a given emulsion relative to the measurements made with a particular microscope.

If z units of the fine-focusing scale, and therefore cz microns, is the vertical projection of an α-particle track measured in the processed emulsion, then the corresponding distance in the emulsion during exposure is $y = khcz$, where k is the shrinkage factor and h is a correction factor for the difference in the refractive indices of the immersion oil and the gelatine of the emulsion. Putting $k' = khc$, then $y = k'z$, where k' is the ~~shrinkage~~ depth conversion factor relative to the microscope used. The range of an α-particle track can now be found from $R = (x^2 + k'^2z^2)^{\frac{1}{2}}$, k'

being determined from measurements made on a hundred tracks produced in the emulsion by monoenergetic α-particles.

From such measurements, a graph of z^2 against x^2 was plotted, the points lying along a straight line of slope $-(1/k'^2)$. In practice R is not strictly constant due to the straggling of the ranges and errors in measurement, and the best straight line through the points was found by the Gaussian method of least squares. The point at which this straight line cuts the x-axis is the square of the mean range of the tracks considered. ~~A series of~~ Straight lines were then drawn parallel to this best straight line representing ranges at intervals of 0.5 micron (see Figure 5). Therefore, to find the range of an α-particle track, the horizontal projection (in microns) and the vertical projection (in units of the fine-focusing scale) were squared and plotted, then the range (in microns) was read off. The ranges were read to the nearest 0.1 micron, although this does not represent the true accuracy of the ranges since errors will have been introduced in the initial measurements of x and z.

This calibration was made for every plate examined; a hundred randomly selected ^{228}Th α-particle tracks being used in both investigations.

One further point must be considered. Under

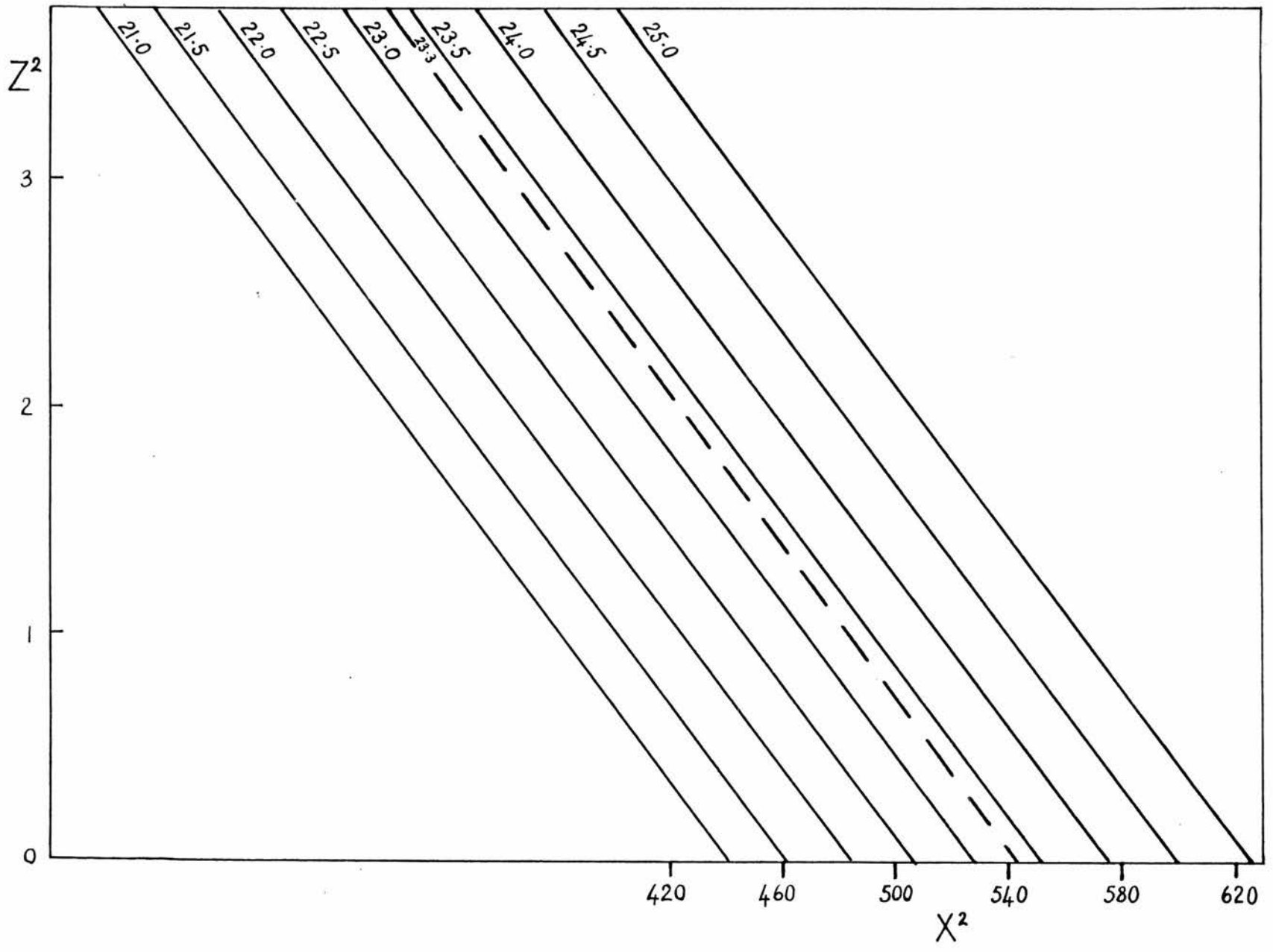

FIGURE 5. FAMILY OF LINES USED IN THE DETERMINATION OF TRACK LENGTHS.

The dotted line was obtained from a least squares calculation.

working conditions the thickness of the emulsion may not be constant in time. Its variation with the relative humidity of the surrounding air has been investigated by Martin (1949). In the present work, day to day variations of up to 5% were observed. In order to avoid the introduction of errors due to this variation, the following precaution was taken. The overall thickness of the emulsion was measured, at a fixed reference point on the plate, at the beginning and the end of each session of observation. By making the reasonable assumption that the variation, if any, was uniform during a session, the total thickness of the emulsion was found at the time of measuring every vertical projection. The relative projections were then expressed relative to a "standard" overall thickness, giving the values of z used in the range determinations.

The ranges of the α-particle tracks as estimated showed no significant variation with steepness. From Plate 1 of the ^{228}Th investigation, four samples, each of a hundred randomly selected α-particle tracks, were considered. The mean range and standard deviation of each sample is given below.

Angle of Dip.	Mean Range.
0° - 10°	$23.44 \pm 0.07\mu$
20° - 30°	$23.46 \pm 0.08\mu$
40° - 50°	$23.46 \pm 0.08\mu$
$\sim 60^{\circ}$	$23.43 \pm 0.11\mu$

III.2. Internal Conversion Electron Measurements.

III.2.1. Introduction.

From the point of view of nuclear emulsion spectrometry, the most important attribute of the conversion electron spectrum is its discreteness. Each monoenergetic group of electrons produces its characteristic population of tracks, and the total population of the plate is the superposition of these populations. In this work the total population is inferred from a random sample and an attempt is then made to resolve this into its component populations. Thus the population of tracks produced by a monoenergetic group of electrons is of fundamental importance.

When a rapidly moving electron passes through matter it can lose energy in two ways. (1) It transfers energy directly to an atom of the surrounding material by exciting or ionising the atom, that is, by inelastic collisions. (2) The electron is deflected in the field of an atom and emits radiation ("Bremsstrahlung"). The formula given by Bethe and

Heitler (1934) enables the relative importance of these two processes to be calculated.

$$\frac{\left(\frac{d\bar{E}}{dR}\right)_{rad.}}{\left(\frac{d\bar{E}}{dR}\right)_{coll.}} \approx \frac{EZ}{816,000}$$

This refers to an electron of energy E keV moving in a material of atomic number Z. $d\bar{E}$ is the average energy loss in moving a distance dR. For electrons of energy approximately 100 keV, the most energetic electrons observed in the present investigations, and for silver, the heaviest atom in the emulsion, this ratio is about 1/160. Therefore, the loss of electron energy is due principally to inelastic collisions.

The average energy transferred in an inelastic collision is of the order of 20 eV, and therefore an electron of initial energy 100 keV will suffer about 5,000 such collisions. For such a process, made up of a large number of small additive contributions, most characteristics of the overall process are distributed normally. This average energy loss may differ considerably from the actual amount of energy lost in any collision and large single-energy transfers do occur. The effect of these large energy transfers is to skew the distribution of the characteristics, but since a large energy transfer is much less probable than a small one, the straggling so introduced is not very large. A larger degree of straggling results

from the radiation losses, since the probability is roughly the same for the emission of quanta of all energies up to very nearly that of the electron. Therefore, although the total energy transferred is governed principally by the inelastic collisions, phenomena which depend upon the rare large single-energy transfers are governed by "Bremsstrahlung".

The tracks produced by electrons of initial energy of roughly 20 keV - 350 keV can be described by their ranges or total grain numbers. The first experimental investigations of the populations of tracks produced in nuclear emulsions by monoenergetic groups of electrons were made by Zajac and Ross (1949). They measured both the ranges and grain numbers of tracks produced by electrons of energies 30, 40, 50, 60, 80, 100, 147, 200 and 250 keV. The mean range for each energy was found to be in good agreement with the value predicted by collision theory. The mean range versus energy relation which they derived is shown in Figure 6. The standard deviation of each range and grain number distribution is about 20% of the mean, a result in agreement with that obtained by Williams (1930) from measurements of the ranges of 20 keV electron tracks in a cloud chamber. Although the number of tracks (25 - 50) considered at each energy was too small to allow an exact analysis of the line shape,

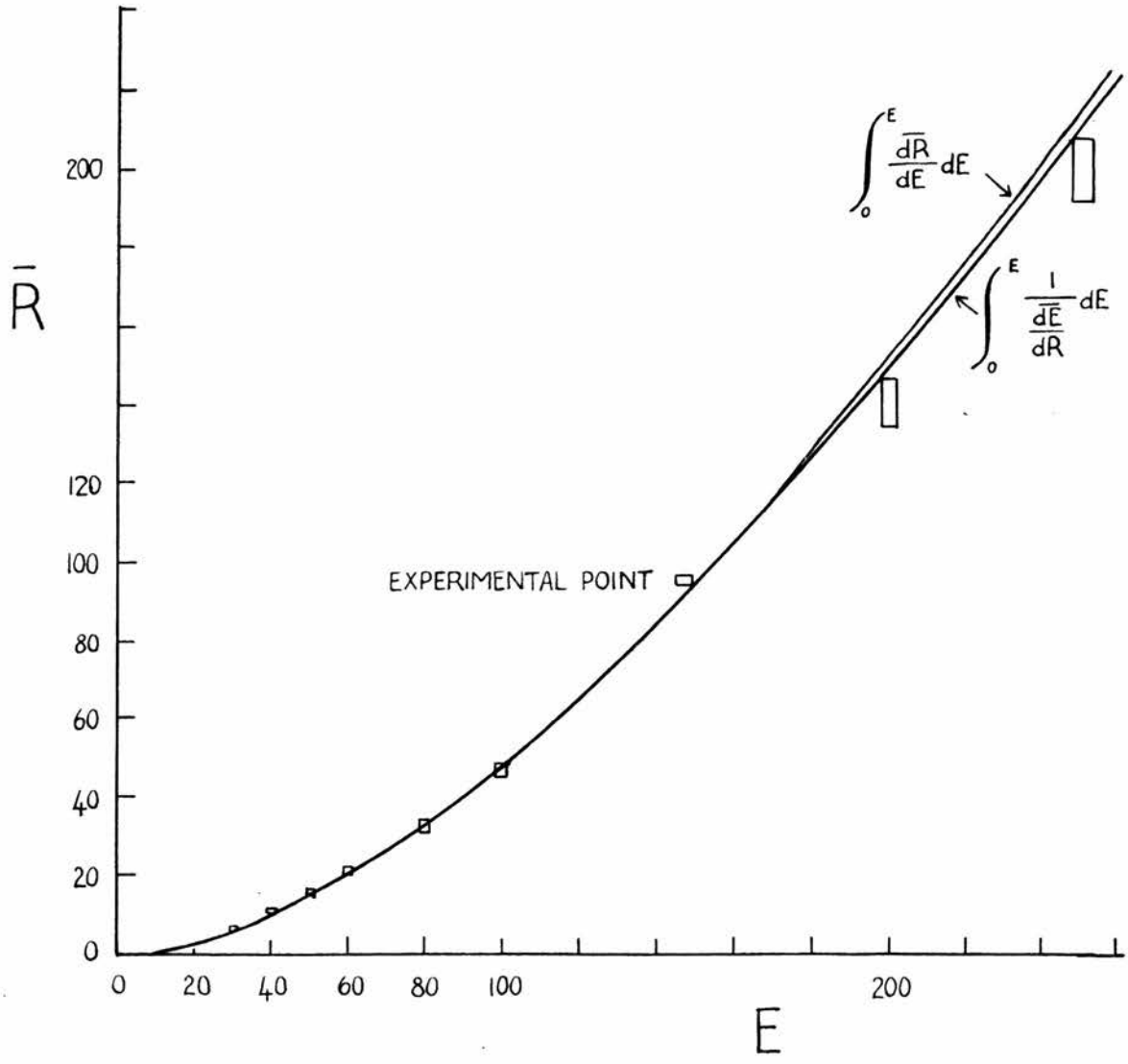

FIGURE 6. MEAN RANGE VERSUS ENERGY FOR MONOENERGETIC GROUPS OF ELECTRONS. CALIBRATION OF ZAJAC AND ROSS (1949). The figure shows the observed mean range in microns ($\bar{R}$) plotted against energy in keV (E) and the curves obtained by integrating $1/\frac{d\bar{E}}{dR}$ and $\frac{d\bar{R}}{dE}$. The correct quantity to integrate is $\frac{d\bar{R}}{dE}$ which refers to the mean distance $d\bar{R}$ gone by particles all of which lose energy dE. $1/\frac{d\bar{E}}{dR}$ refers to the mean energy $d\bar{E}$ lost by particles all of which move a distance dR. The numerical difference arises because of the straggling of the energy losses and the non-linearity of $\bar{R}(E)$. The correctly calculated mean range versus energy curve gives a slightly poorer fit with the observations than does the incorrectly calculated one.

Zajac concluded that the determination of energy on the basis of the grain counts would be as good as that obtained from the range measurements. The results obtained by Bayman (1954) are in agreement with this conclusion.

III.2.2. Experimental Method.

The estimation of the range of an electron track was made by the same method as that used for α-particle ranges. The electron track was broken up into straight-line segments, the length of each segment estimated, and the range found by the addition of these lengths. This method is very laborious especially as almost all electron tracks consisted of at least ten segments and some had to be broken up into more than twenty segments. A more serious obstacle encountered in range measurements is due to the fact that some electron tracks are very tortuous at the low energy end. This made an accurate determination of range very difficult and sometimes impossible. Obviously, the errors so introduced will be more serious for the tracks of electrons at the low energy end of the spectrum than for those of higher energy electrons. The occasional occurrence of branching in an electron track does not introduce a serious error. Branching is a result of the incident electron, during the rare large ionising collisions, transferring sufficient energy to

an atomic electron to enable the latter to produce a track of its own. Because of the indistinguishability of electrons and because the incident electron may transfer any fraction of its energy to the atomic electron it is impossible to say which of the resulting tracks is due to the primary electron. In such cases, following the conventional procedure, the longer of the two tracks was taken to be that of the incident electron. Since the contribution to the line width from the rare collisions, in which branch tracks are produced, is very small compared with the straggling resulting from the radiation losses, it is not worth taking account of the shorter track. In grain counting, the grains in both branches were taken into account, the idea being that the total grain number should depend on the total ionisation, that is both primary and secondary, in analogy with proportional counter methods.

In a well-developed emulsion the individual grains are well defined except at the low energy end of the track, where the grains tend to be formed in clumps. This presented a difficulty similar to that found in the range measurements, but it was easier to estimate the number of grains involved in such a clump than to measure the length of this portion of the track. This estimation consisted of assessing how many grains

could be fitted into the clump, the grain size being taken from the high energy part of the track. In an attempt to avoid the introduction of systematic errors due to the possible preference of the observer for certain numbers, the grain count in each straight-line segment of the track was noted and the total grain number found only after the completion of the counting. All measurements were made from the low energy end of the track.

One drawback affecting the determination of energy from grain number measurements is that of calibration. The ranges of electrons in the emulsion depend only on the chemical composition of the emulsion (provided the moisture content is controlled during exposure), whereas the grain numbers depend upon the processing as well. Earlier investigators have remarked that this dependence on processing would necessitate a separate calibration for each plate. The results obtained in the present work show that, provided reasonable care was taken in the processing, the mean grain number for a monoenergetic group of electrons is the same for different plates (see Figures 7 and 8). However, this constancy is unlikely to hold for grain counts made by different observers. Therefore, if grain numbers are to be used in energy determinations, a calibration must be made for each

investigation by the observer concerned. For the ^{232}Th investigation the calibration was obtained from the ranges and grain numbers of 142 electrons (see Figure 10). In the case of ^{228}Th the method of calibration was somewhat different, and the discussion of this will be left until Chapter V.

III.3. Smoothing.

In order to obtain accurate estimates of the most probable values of the ranges and grain numbers the raw histograms were smoothed. From a consideration of the histograms it was decided to use, with one exception, a seven term smoothing formula

$$u'_0 = \frac{1}{21}\left\{7u_0 + 6(u_1 + u_{-1}) + 3(u_2 + u_{-2}) - 2(u_3 + u_{-3})\right\}$$

The exception was the number versus range histogram of the ^{232}Th conversion electrons, to which the Spencer twenty-one term summation formula was applied. The results obtained were satisfactory.

Chapter IV.

RESULTS FROM THE INVESTIGATION OF THE α-DISINTEGRATION OF ^{232}Th.

The results to be reported here were obtained from two plates which had been impregnated at different times, with different solutions. Plate 1, after observations had been made on 1,500 events, was found to be partially desensitized and this resulted in a serious deficiency of conversion electron tracks. Plate 2 yielded satisfactory numbers of conversion electron tracks. Because of this, the following interpretation is based on the data obtained from Plate 2, although the results from Plate 1, with the exception of the conversion electron intensity, are shown to be consistent with the conclusions reached. The main reason for the inclusion of the results obtained from Plate 1 is to show the close agreement between the electron track grain number distributions in the two plates.

IV.1. Experimental Results.

Measurements were made on the tracks of α-particles and conversion electrons in events which showed a single α-particle track with a range not greater than about 27μ.

Plate 1.

In this plate 1,500 such events were considered, of which 249 showed a conversion electron track. From the frequency distribution of the α-particle ranges, the presence of three groups was observed (see Figure 11). These groups, with mean ranges of 15.3 μ, 18.6 μ and 23.2 μ, were attributed to the α-disintegration of ^{232}Th, ^{230}Th and ^{228}Th respectively. The resolution of these groups is not complete.

Since it was necessary, for the purposes of this investigation, to make a reasonably accurate estimate of the number of events associated with the disintegration of ^{232}Th, some method of separating such events from those of ^{230}Th had to be found. The method adopted involved the use of results obtained by Jarvis and Ross (1951) from an emulsion study of the disintegration of ^{230}Th, and will be described in some detail when the results from Plate 2 are discussed. On the basis of this method, 1,200 α-particle tracks, of which 200 showed a conversion electron track, were considered as having been produced in the disintegration of ^{232}Th. The resulting conversion electron intensity of 17% is much lower than the values given by Albouy (20%) and by Dunlavey (24 ± 3%).

Because of this discrepancy, a check was made on the number of conversion electrons associated with the

^{228}Th α-particles. From Figure 11 140 α-particle tracks were attributed to this disintegration, and of these only 23 had an associated electron track. Comparison of this with the expected number of 36 indicated that the emulsion had not recorded all the conversion electrons which had been emitted. Various other plates were impregnated with freshly prepared active solutions and different development times were used, but the electron intensity found was still too low. Finally, the activity of the impregnating solution was reduced by a factor of 100 and the observed electron intensity increased to the expected value (Plate 2). This suggested that the desensitization of the emulsion may have been due to a chemical action of the thorium salt on the gelatin.

Although the desensitization resulted in a low intensity of conversion electron tracks, it did not seriously affect the grain numbers of the tracks which were formed. The grain number distribution is shown in Figure 7 and the most probable grain numbers of the two groups are consistent with the respective values found from the distribution in Plate 2.

Plate 2.

Because of the very low activity of the impregnating solution only 1,000 events were considered in this plate. 240 of these events showed an associated

conversion electron track. As before, the groups resulting from the disintegrations of ^{232}Th, ^{230}Th and ^{228}Th were observed in the α-particle range distribution (see Figure 12a).

From a study of the ^{230}Th disintegration, Jarvis and Ross found a conversion electron intensity of 24%. They also obtained the frequency distribution for 4,687 α-particle ranges, with a mean range of 18.63 ± 0.01μ. The maximum height of the distribution was just less than 13% of the total number of events considered.

The maximum height of this group, as observed in the present plate, is 12 and, therefore, the number of events in the group was estimated as 100. As in Plate 1, it was considered that about 140 α-particle tracks were associated with the ^{228}Th disintegration and, therefore, 760 events were interpreted as belonging to ^{232}Th. Since the number of conversion electrons associated with the ^{228}Th α-particles gave an intensity consistent with the accepted value of 27%, it was assumed that 24 of the observed conversion electrons would belong to ^{230}Th and, therefore, 180 were assigned to ^{232}Th. This led to an electron intensity of 24 ± 3% for the ^{232}Th disintegration.

One further point must be considered. In order to obtain the range and grain number distribution for the electrons emitted following the disintegration of

^{232}Th, it was necessary to identify such events with certainty. This identification could not be made from a consideration of the electron tracks themselves, since the energies of the ^{232}Th electrons and those of ^{230}Th are too close together to permit resolution. The identification, therefore, had to be performed on the basis of the associated α-particle ranges. The results of Jarvis and Ross indicate that, of the 824 α-particle tracks with associated electrons considered, 5% had ranges less than 17μ. This percentage, when related to the number of such events observed in Plate 2, led to the conclusion that only 1 or 2 ^{230}Th events should have ranges less than 17μ. The distribution shown in Figure 12b is consistent with this conclusion.

IV.2. <u>The Conversion Electron Spectrum.</u>

Figure 9 shows a histogram and smoothed curve of the ranges of 142 conversion electrons associated with the α-particles emitted in the ^{232}Th disintegration. The smoothed curve indicates the existence of two groups with mean ranges of 11.4μ and 17.5μ. From the mean range versus energy relation of Zajac and Ross (1949) shown in Figure 6, the energies associated with these ranges were estimated as 42.0 keV and 54.5 keV respectively. The groups were therefore interpreted

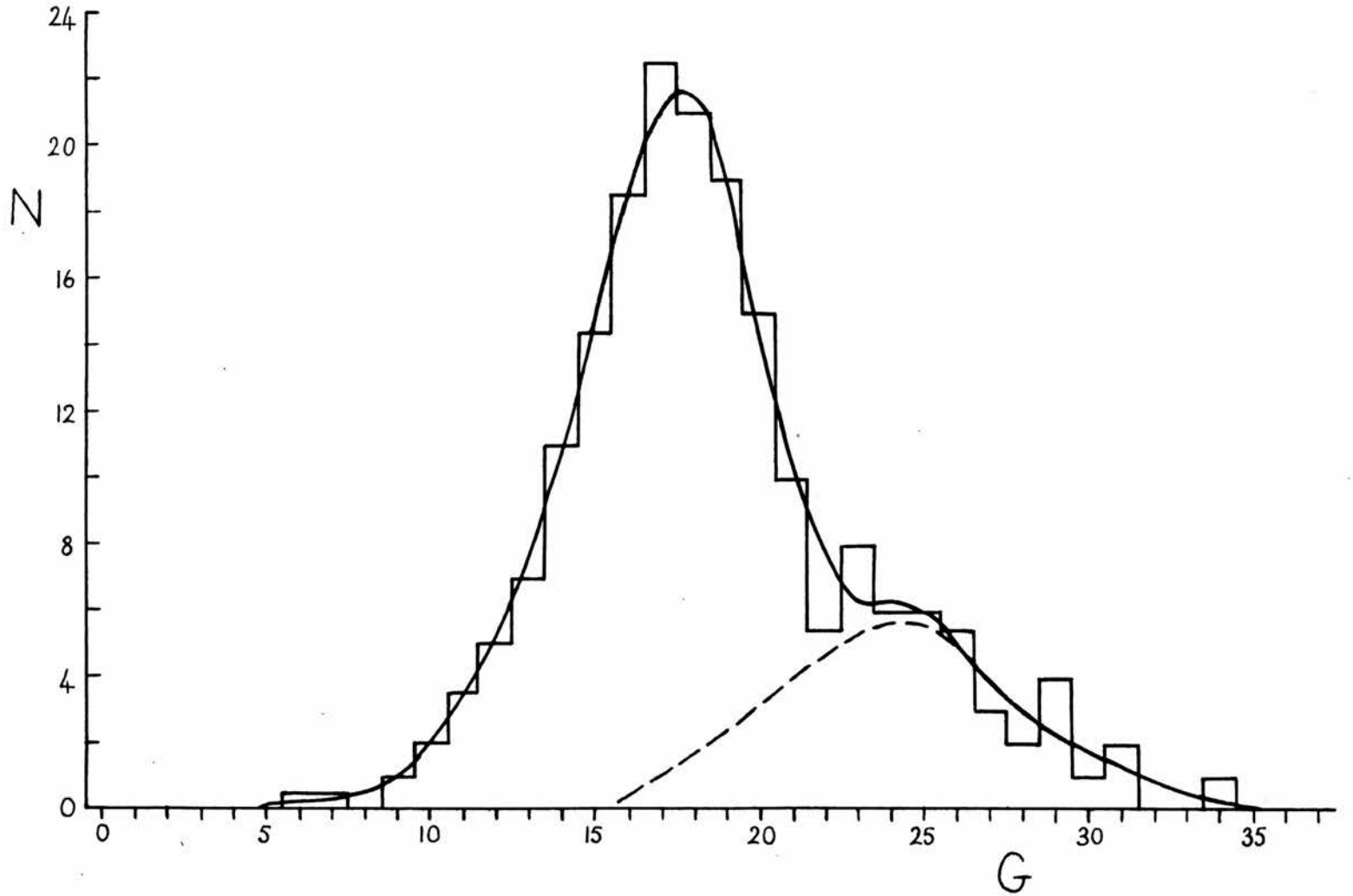

FIGURE 7. FREQUENCY (N) VERSUS GRAIN NUMBER (G) OF CONVERSION ELECTRONS EMITTED FOLLOWING THE ^{232}Th DISINTEGRATION (PLATE 1).

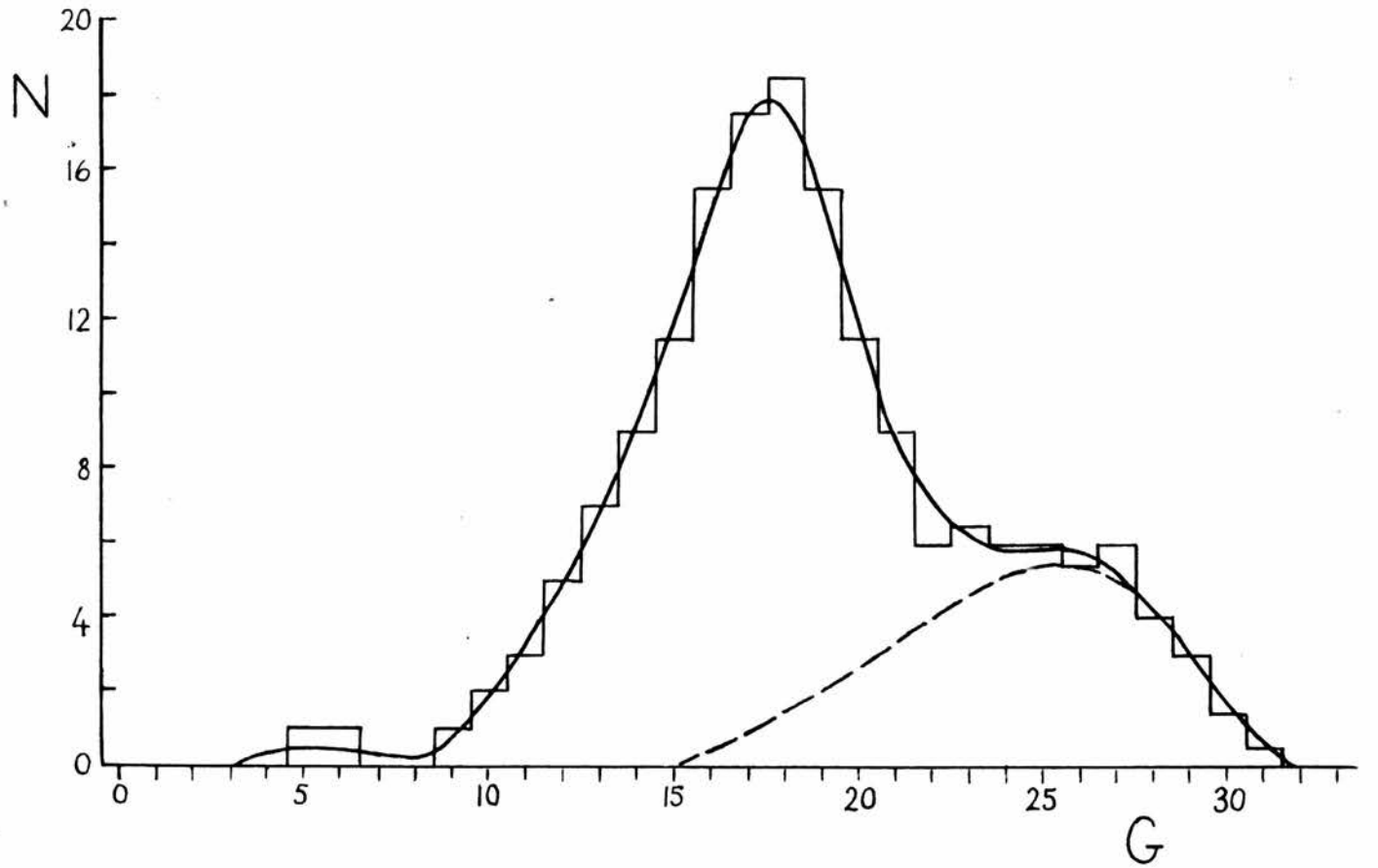

FIGURE 8. FREQUENCY (N) VERSUS GRAIN NUMBER (G) OF CONVERSION ELECTRONS EMITTED FOLLOWING THE ^{232}Th DISINTEGRATION (PLATE 2).

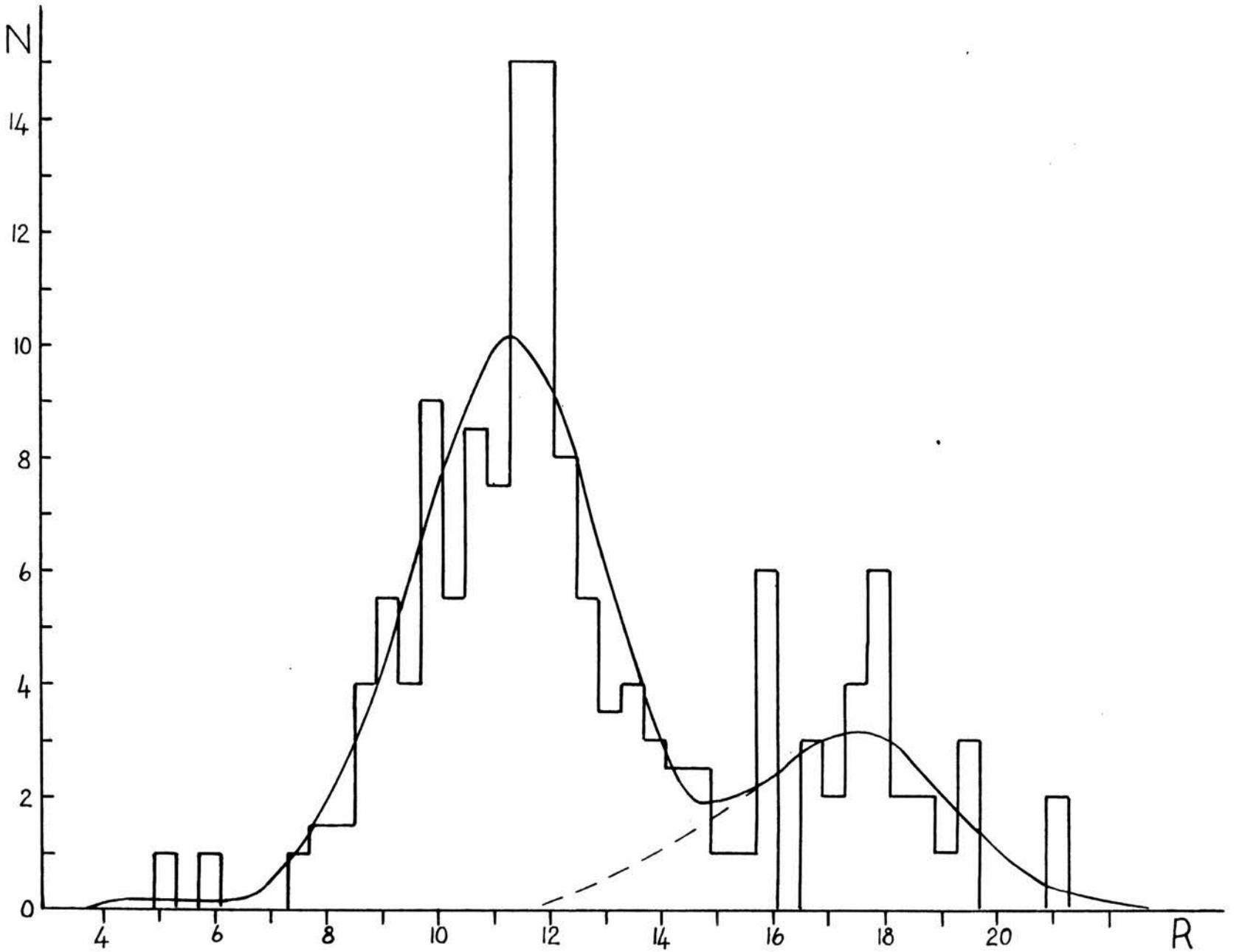

FIGURE 9. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF CONVERSION ELECTRONS EMITTED FOLLOWING THE ^{232}Th DISINTEGRATION. (PLATE 2).

as representing electrons emitted from the L- and M + N-shells of radium. The smooth curve was resolved into its component groups by the following method. It was assumed that the low energy side of the main peak was not seriously affected by the higher energy group. On this assumption various parameters were calculated for the lower energy line. The half-widths at half, quarter and three-quarters of the maximum height were estimated as 18%, 28% and 11% of the mean range respectively. From these percentages, the three half-widths for a mean range of 17.5 μ were calculated. The results given by Jarvis and Ross (1951) and later those obtained by Rosenblum, Valadares and Guillot (1952; 1954) from a study of ^{228}Th indicate that the ratio of L-conversion electrons to M + N-conversion electrons is about 3:1. Using the calculated half widths and this ratio, the low energy side of the higher energy line was plotted, as shown by the broken line. This resolution led to mean ranges, with standard errors, for the two groups of 11.25 $\pm$ 0.25 μ and 17.5 $\pm$ 0.4 μ . The standard deviations of the distributions about their means were calculated as 23% and 14% of the mean ranges respectively.

From the relation of Zajac and Ross, the respective energies were estimated as 42.0 $\pm$ 0.5 keV and 54.5 $\pm$ 0.7 keV. These electrons were assumed to have

been emitted in an E2 transition from the first excited level of ^{228}Ra. The internal conversion coefficients for the L_I, L_{II} and L_{III} sub-shells were calculated* for E2 radiation of 60 keV as 2, 51 and 40 respectively. From these values it follows that the L-group was due mainly to conversion in the L_{II} and L_{III} sub-shells, which have term values of 18.48 keV and 15.44 keV (Cauchois, 1952). The M + N-group probably arose mainly from conversion in the M_I, M_{II} and M_{III} sub-shells, which have term values between 4.8 keV and 3.8 keV. Therefore, for the purposes of this investigation, the effective binding energies of electrons in the L- and M-shells were taken as 17.0 keV and 4.3 keV respectively. The energy of the transition was then given by 42.0 + 17.0 = 59 keV from the L-conversion group and 54.5 + 4.3 = 58.8 keV from the M-conversion group. Its value was therefore taken as 59 $\pm$ 1 keV.

The grain number distribution of 173 conversion electrons (see Figure 8), like the range distribution, shows the existence of two groups. The peak grain numbers of these groups are $17\frac{1}{2}$ and 25. Proceeding in the same way as before, this distribution was resolved into its component parts. The half-widths of

* Calculations based on the tables of conversion coefficients given by Rose, Goertzel and Swift (Private communication).

the main peak were estimated as 18%, 30% and 12%, in good agreement with the corresponding values from the range distribution. The most probable grain numbers of the resolved groups were taken as 17 and 25. The respective standard deviations of these groups were estimated as 20% and 13%. From the grain number versus range calibration (see Figure 10) the ranges associated with the two groups were found to be 11.3 μ and 17.8 μ respectively, which are consistent with the values previously determined. The grain number distribution obtained from Plate 1 is shown in Figure 7. The peak grain numbers are 18 and 24 and the half-widths calculated from the low energy side of the main peak are 18%, 30% and 12%. The most probable grain numbers of the resolved groups were estimated as 17 and $24\frac{1}{2}$, in good agreement with the values obtained from Plate 1. The standard deviations of the component distributions were calculated to be 17% and 15% respectively.

These results indicate that, provided the grain numbers are properly calibrated, the energies deduced from the range and grain number measurements are consistent. They also show that the grain number versus range calibration constructed from the observations made on one plate can be applied successfully to those made on another plate.

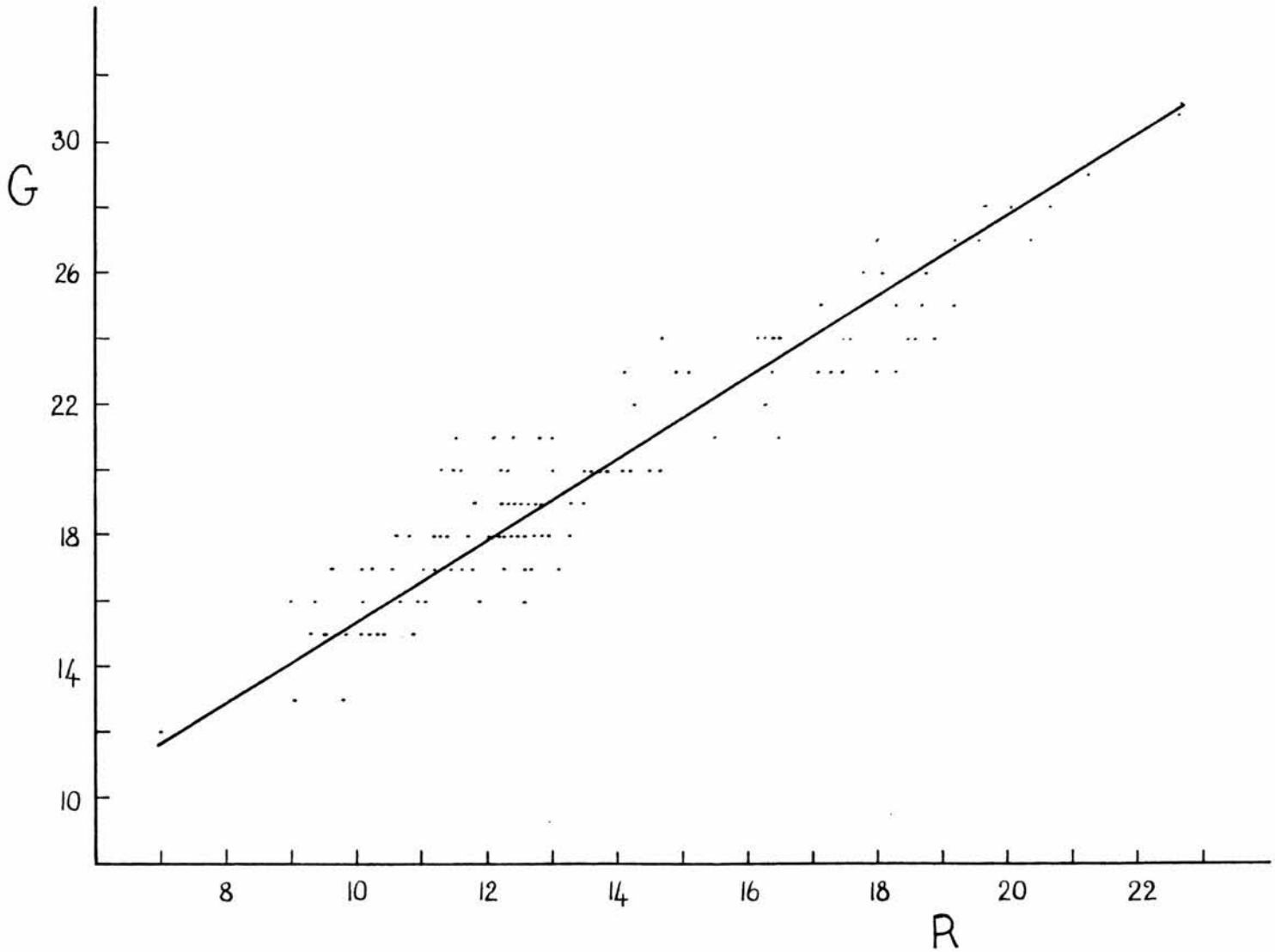

FIGURE 10. GRAIN NUMBER (G) VERSUS RANGE IN MICRONS (R) FOR ELECTRON TRACKS - CALIBRATION (PLATE 2).

As both Albouy and Dunlavey pointed out, the intensity of the conversion electrons represents a lower limit of the intensity of the α-particle feed to the excited level. No direct measurements have been made on the α-particle feeds but by using the internal conversion coefficients it was possible to estimate the intensity of the γ-radiation. The ratio of the L-conversion electrons to the M + N-conversion electrons was 3:1 and, therefore, the intensity of the L-conversion electrons was approximately 18% of the α-emission. Using the total conversion coefficient for the L-shell of 93 led to a γ-ray intensity of 0.2%.

No electron tracks were observed which could have been attributed to the de-excitation of a higher excited level in ^{228}Ra.

IV.3. The Auger Electrons.

Vacancies are produced in the electron shells by the emission of conversion electrons during the de-excitation of the nucleus. When such a vacancy is filled by an electron from a higher shell the excess energy is carried away either by X-radiation or by an Auger electron. In the present case vacancies were produced in the L- and M-shells of ^{228}Ra. The mean energy of the L X-radiation is about 14 keV and, therefore, the energy of an Auger electron from the M-shell

would be about 9 keV. This would give rise to an electron track with a range of 1 μ. Such a track, consisting of 1 or 2 grains, would very easily be missed or attributed to background grains and it could be misinterpreted as the initial grain of the longer conversion electron track which must always be present. Longer tracks of Auger electrons from the outer shells, or from more energetic L X-radiation, would have a maximum range of about 2 μ and consist of up to 4 grains, but the identification would still be difficult. The Auger electrons which result from the filling of vacancies in the M-shell need not be considered due to their very low energies.

It has been calculated, on the basis of results obtained in this laboratory from a study of ^{233}U, that only 10% of the Auger electrons are visible, as such, in the emulsion. From the 180 conversion electron tracks observed in Plate 2, 7 showed associated tracks which were interpreted as being due to an Auger electron. The number of vacancies produced in the L-shell was estimated as 135 and, by assuming that the intensities of the X-radiation and the Auger electrons were in the ratio of $\frac{0.4}{0.6}$ (estimated from the Table of Fluorescent Yields given by Burhop, 1952), it was expected that about 80 Auger electrons should have been emitted. The observed number results in a visibility

factor of 9%, in good agreement with the value previously calculated.

IV.4. The α-radiation.

The α-particle range distributions are shown in Figures 11, 12a, b, c. The half-widths at half-maximum of the main peaks were 0.6 μ. From the distributions obtained from Plate 2 the mean ranges, with standard errors, of the lines in the ^{232}Th α-particle spectrum were estimated.

For all α-particles the mean range was found to be 15.30 $\pm$ 0.02 μ. The α-particles were then separated into two groups, those with associated conversion electrons, and those without conversion electrons. The mean ranges of these groups were 15.07 $\pm$ 0.05 μ and 15.38 $\pm$ 0.04 μ respectively. In order to determine whether or not these values could have been obtained by random selection from a single population of α-particle tracks, a Student's "t-test" was applied to the differences. For the difference of 0.23 μ between the mean ranges of all α-particles and that of those with associated conversion electrons, the value of t was 3.94. Since, with a value of t = 2.58, the probability of obtaining such a difference by chance was 1 in 100, this difference was taken as highly significant and the mean ranges of 15.30 $\pm$ 0.02 μ

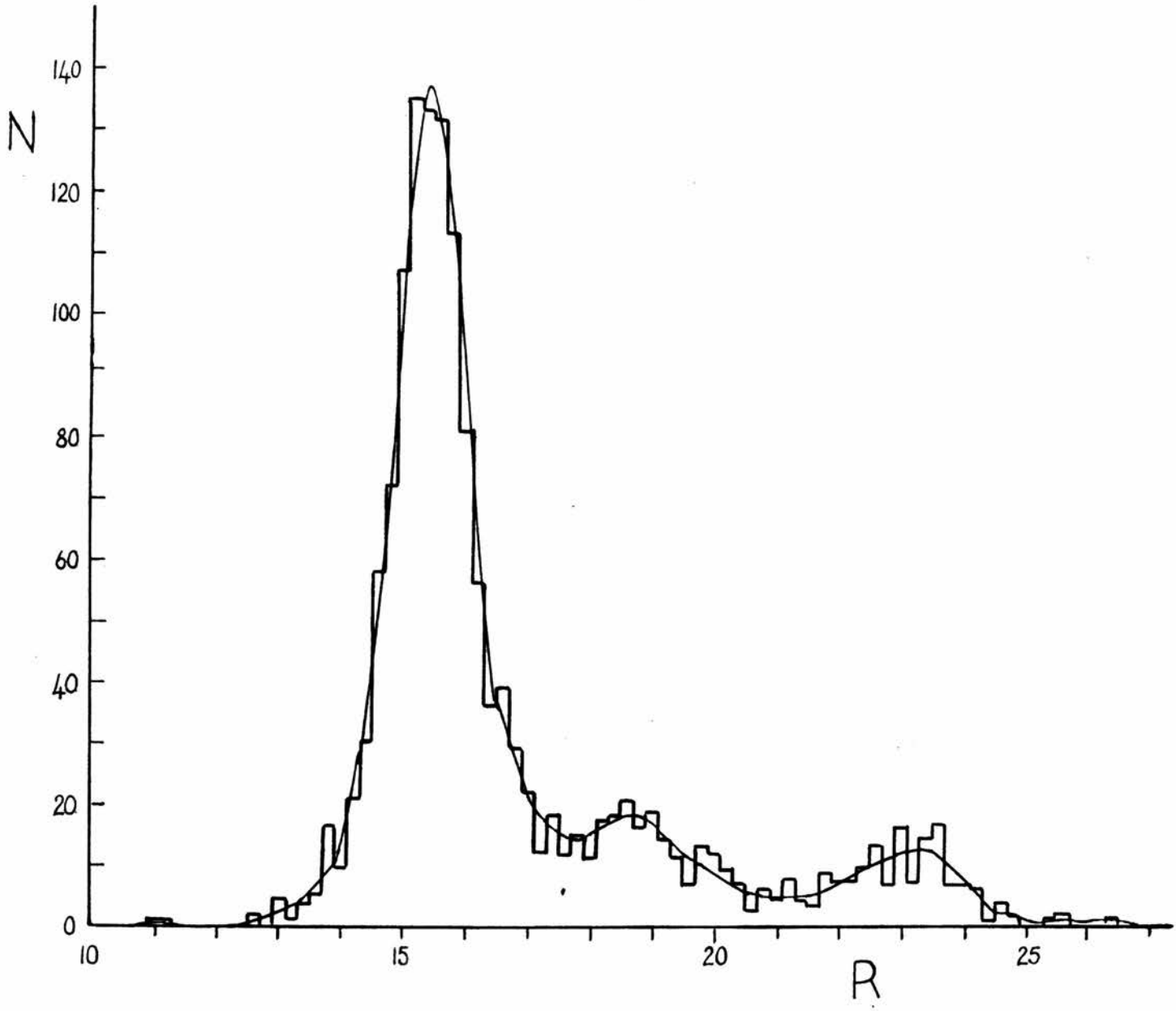

FIGURE 11. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF "ALL" α-PARTICLES (PLATE 1).

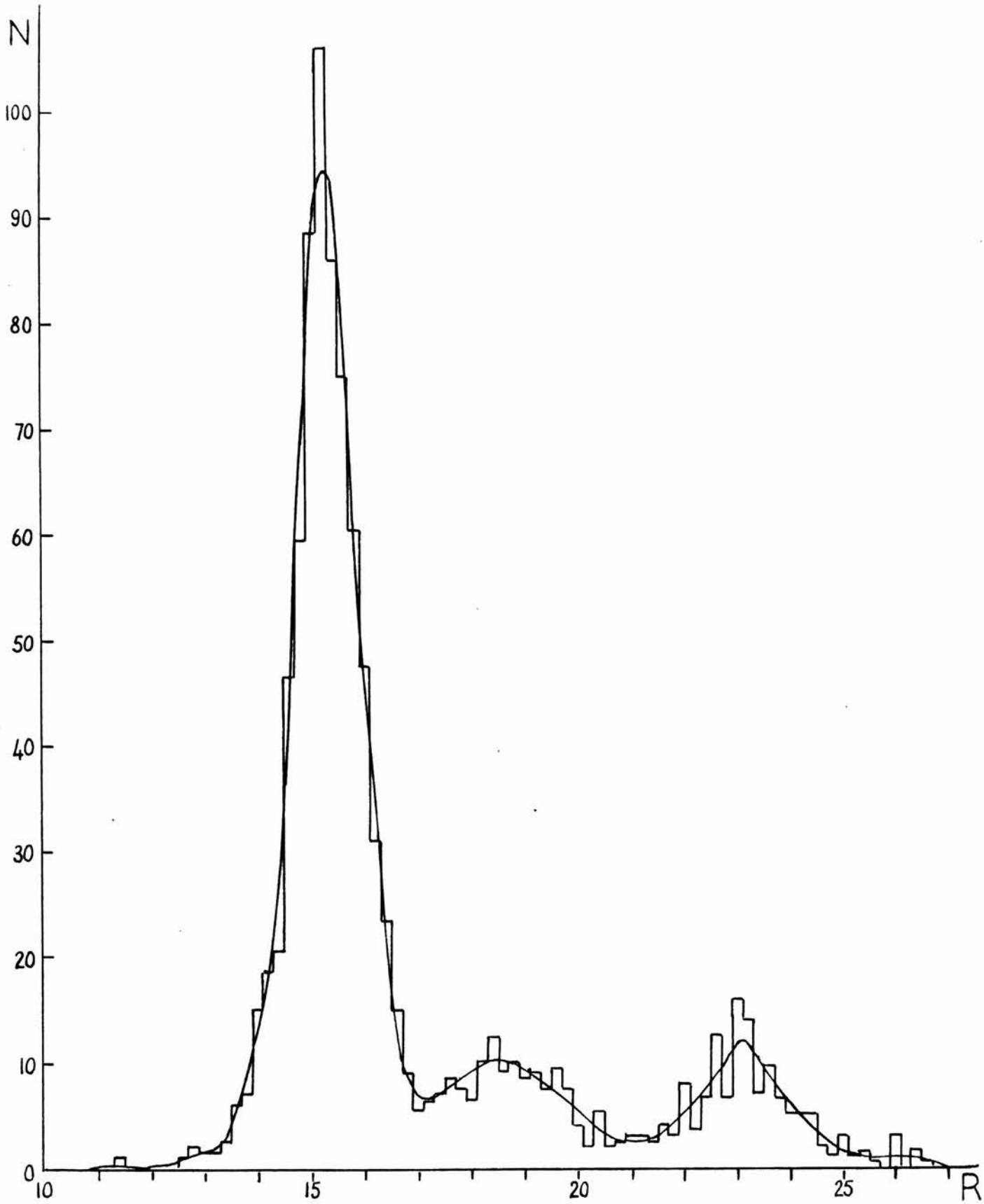

FIGURE 12a. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF "ALL" α-PARTICLES (PLATE 2).

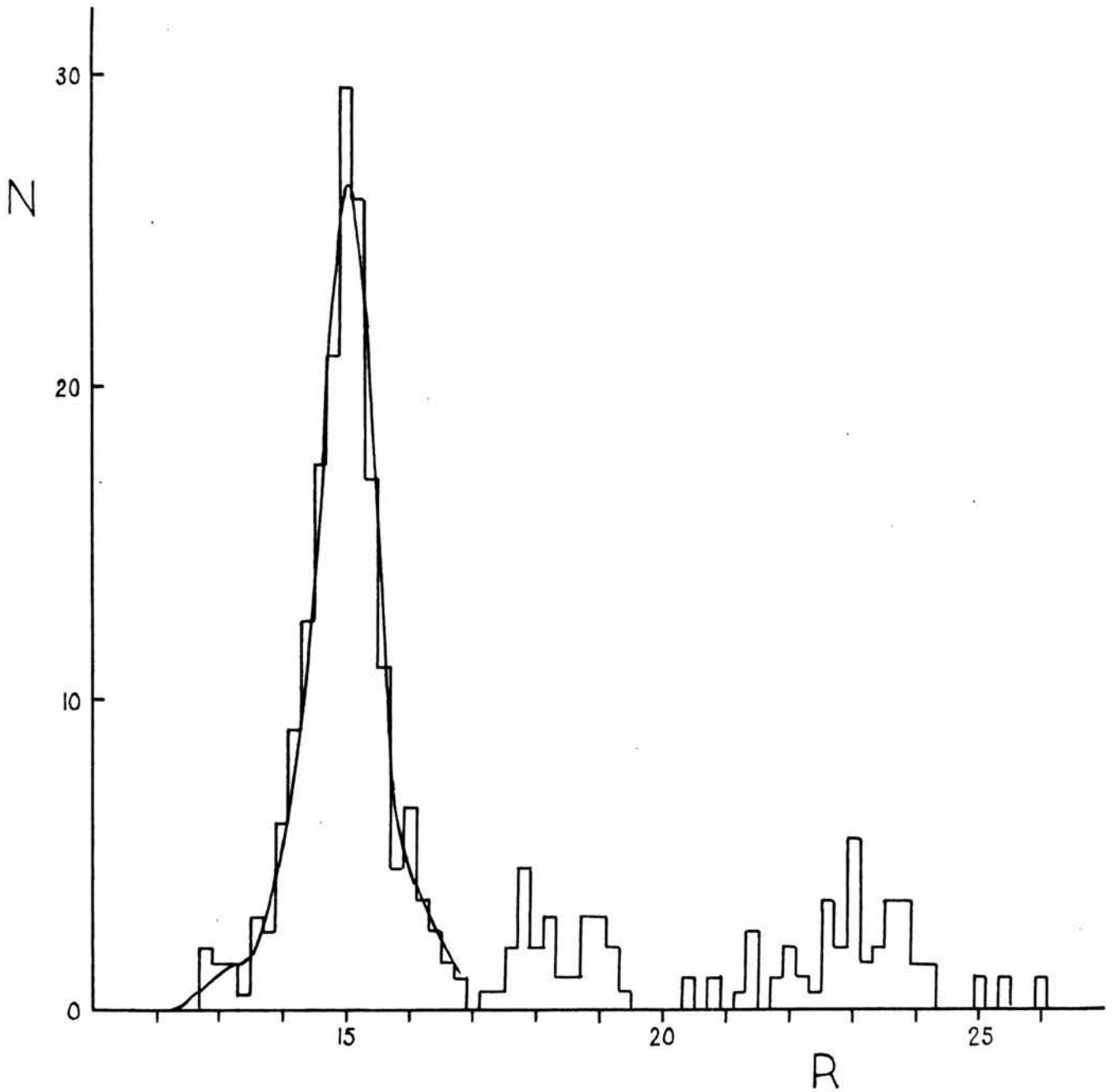

FIGURE 12b. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF α-PARTICLES WITH ASSOCIATED CONVERSION ELECTRONS (PLATE 2).

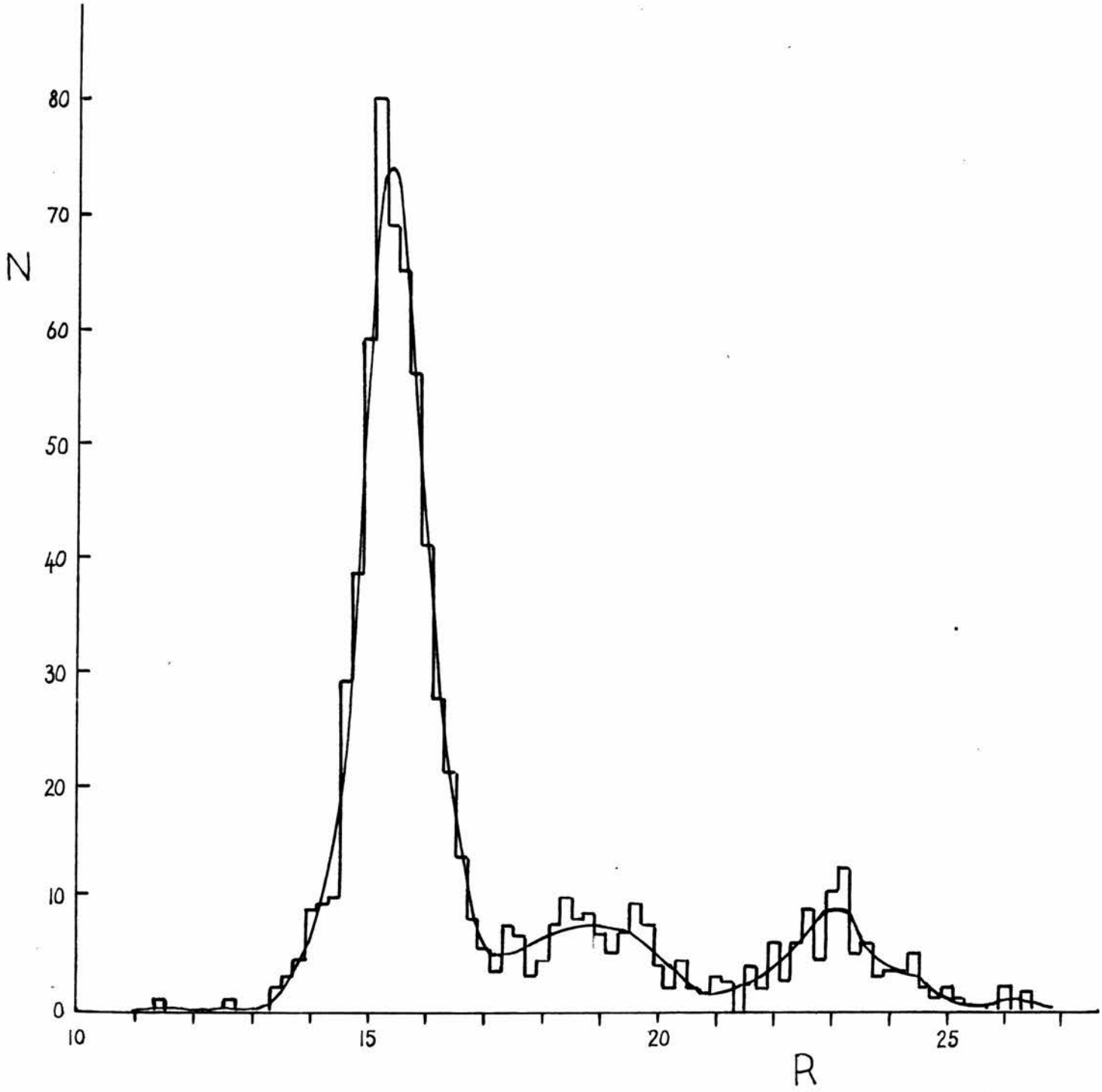

FIGURE 12c. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF α-PARTICLES WITH NO ASSOCIATED CONVERSION ELECTRONS (PLATE 2).

and 15.07 ± 0.05 μ were considered to represent two α-populations. The difference of 0.08 μ led to a t-value of 1.82 which represented a chance probability of about 1 in 20. When the difference of 0.31 μ was considered the value of t was found to be 4.40.

The mean ranges of the α-particles feeding the ground state and the first excited level of ^{228}Ra were therefore taken as 15.38 ± 0.04 μ and 15.07 ± 0.05 μ respectively. The difference of 0.31 ± 0.06 μ corresponded to a difference of 60 ± 12 keV between the kinetic energies of the α-particles. This energy difference is consistent with the nuclear energy difference deduced from the conversion electron spectrum and its interpretation in terms of conversion in the L- and M-shells of ^{228}Ra.

The α-particle tracks observed in Plate 1 were not separated into component groups because of the loss of sensitivity in the emulsion. Some α-particles which were not associated with a conversion electron track did, in fact, feed the first excited level of ^{228}Ra, and their inclusion in the α-line leading to the ground state would have introduced errors in the determination of the energy difference. However, the mean range of all the ^{232}Th α-particles found in this plate was calculated as 15.33 ± 0.02 μ and is consistent with that obtained from Plate 2.

The energy difference of 60 ± 12 keV, quoted above, was estimated from the mean range versus energy calibration of Rotblat (1950). Since this calibration was derived from measurements made on α-particle tracks in emulsions which had a different stopping power from those used in the present work, no attempt was made to estimate the absolute energies of the two α-particle lines found in the ^{232}Th spectrum.

IV.5. Conclusion.

The results obtained from this investigation lead to the conclusion that the first excited level of ^{228}Ra lies at 59 ± 1 keV above the ground state and is excited in 24 ± 3% of the α-disintegrations of ^{232}Th. This energy is consistent with the trend exhibited by the energies of the first excited levels in the even mass number isotopes of radium (see Figure 13).

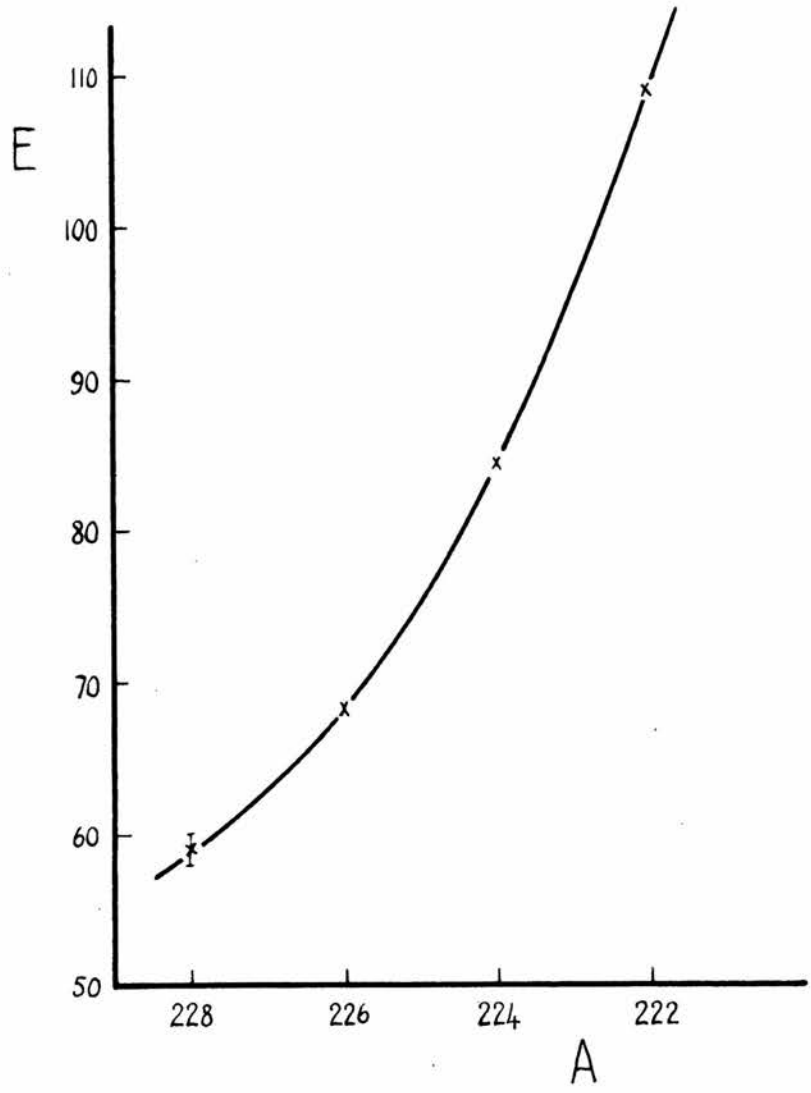

FIGURE 13. ENERGY IN keV OF THE FIRST EXCITED LEVEL (E) AGAINST MASS NUMBER (A).

Plotted for the even mass number isotopes of radium.

Chapter V.

RESULTS FROM THE INVESTIGATION OF THE α-DISINTEGRATION OF ^{228}Th (RADIOTHORIUM).

V.1. Experimental Results.

Plate 1.

In the initial examination of this plate, measurements were made on the tracks of α-particles and conversion electrons in 1,500 events which showed a single α-particle track. From a consideration of the α-particle ranges, all these events were attributed to the disintegration of ^{228}Th. The conversion electron spectrum showed, as well as the electrons which had been emitted in the de-excitation of the first excited level of ^{224}Ra, some electrons which were interpreted as having been emitted in the de-excitation of higher levels. A further 2,000 events were considered but measurements were made only on those in which a conversion electron was exhibited. Finally, another 2,000 events were observed, a count kept of the number of all conversion electron tracks and measurements were made on those in which the α-particle was associated with an electron emitted in the de-excitation of a higher excited level. The advantage of this method of working is that, once the active content of the emulsion has been established,

the number of low intensity events observed can be increased with an economy of effort.

One group of electrons found in this plate was suspect on the grounds of the observed intensity. This group consists of ten electrons with grain numbers between 72 and 84, but of these three were doubtful in so far as the origins of the electron tracks were not certain. On the basis of the level scheme suggested by Asaro, Stephens and Perlman (see Figure 3), these electrons could be interpreted only as having been emitted from the K-shell in a transition between the 217 keV level and the ground state. The intensity of these electrons, 0.13 ± 0.05% of the ^{228}Th α-emission, is, however, too high to agree with E1 radiation from the 217 keV level. If these electrons did occur in this transition, then it is to be expected that the mean range of the associated α-particles would be lower than that of the α-particles feeding the ground state, due to the difference in available energy. While it is not to be expected that the difference in the mean ranges, as observed in this experiment, would give an exact quantitative agreement with the predicted energy difference, it is reasonable to suppose that it would show qualitative agreement. In the present case, however, the ranges of the α-particles, interpreted as feeding the 217

keV level, lay between 23.00 μ and 25.00 μ, with a mean of 23.74 μ. This mean range is about 0.2 μ higher than that calculated for the α-particles feeding the ground state and therefore provided another reason for doubting the above interpretation. It seeme probable that these electrons resulted from a contamination present in the impregnating solution.

Since no α-particle groups were observed which had ranges significantly different from that found for the ^{228}Th α-particles, the most likely contamination is ^{224}Ra, of which the principal α-group has a mean range in emulsion of about 25 μ. The disintegration of ^{224}Ra leaves the daughter nuclei in an excited state with an intensity of 4.6% of the total α-emission. This excited level, 241 keV above the ground state, is de-excited mainly by the emission of conversion electrons. The expected mean range of the α-particles feeding this level is about 23.5 μ which is reasonably close to the observed mean range of 23.74 μ. It is therefore possible that the conversion electrons, interpreted as having followed the disintegration of ^{228}Th, may have followed the disintegration of ^{224}Ra.

A difficulty, which arises from this explanation, is that it appears to be unlikely that α-particles from the disintegration of ^{224}Ra would be observed as

single tracks. Even if the impregnating solution had contained an appreciable amount of ^{224}Ra, the α-disintegration would be followed almost immediately by the α-disintegrations of ^{220}Em (Tn) and ^{216}Po (ThA) and, after a short hold-up at ^{212}Pb, by successive and rapid transformations leading to ^{208}Pb (see Figure 1). Thus, an α-particle emitted in the disintegration of ^{224}Ra was expected to form part of a "star" consisting of three and probably four α-particle tracks. There is, however, one way in which a ^{224}Ra α-particle could appear as a single track. The immediate disintegration product of ^{224}Ra is the ^{220}Em which is able to diffuse freely through the emulsion. Since the half-life of ^{220}Em is short, 54.5 seconds, the magnitude of this diffusion, under normal conditions of exposure, is very small and has a most probable value of 2 - 3 microns. When the emulsion is "wet" it is possible that the magnitude of the diffusion could be very much larger and, therefore, provided the disintegration of ^{224}Ra occurred in such conditions, the emitted α-particles might be observed as single tracks. If this explanation is correct, it is surprising that the mean range of the α-particle tracks should show such good agreement with that expected for tracks formed under strictly controlled conditions. This hypothesis, however, gained some support from the

fact that the intensity of "stars" consisting of four α-particle tracks was higher than would be expected in a plate impregnated with pure ^{228}Th solution.

On the basis of a conversion electron intensity of 4.6%, it was necessary to attribute about 150, of the 5,500 α-particles considered, to the disintegration of ^{224}Ra.

A second impregnation was performed with a solution of ^{228}Th freshly purified as described in Chapter II. The results obtained from the examination of one of the plates so impregnated (Plate 2) differed from those obtained from Plate 1 only in so far as the doubtful group of electrons appeared with a greatly reduced intensity. This suggested that, whether or not the explanation given above is correct, this group, as observed in Plate 1, did not belong to the disintegration of ^{228}Th.

Plate 2.

Measurements were made on the first 500 single α-particle tracks observed and all were attributed to the disintegration of ^{228}Th. The grain numbers of the associated conversion electrons showed no significant difference from those obtained from Plate 1 (see Figures 14 and 15). A further 4,000 events were considered and measurements were made only on those in which the α-particle was associated with a conversion

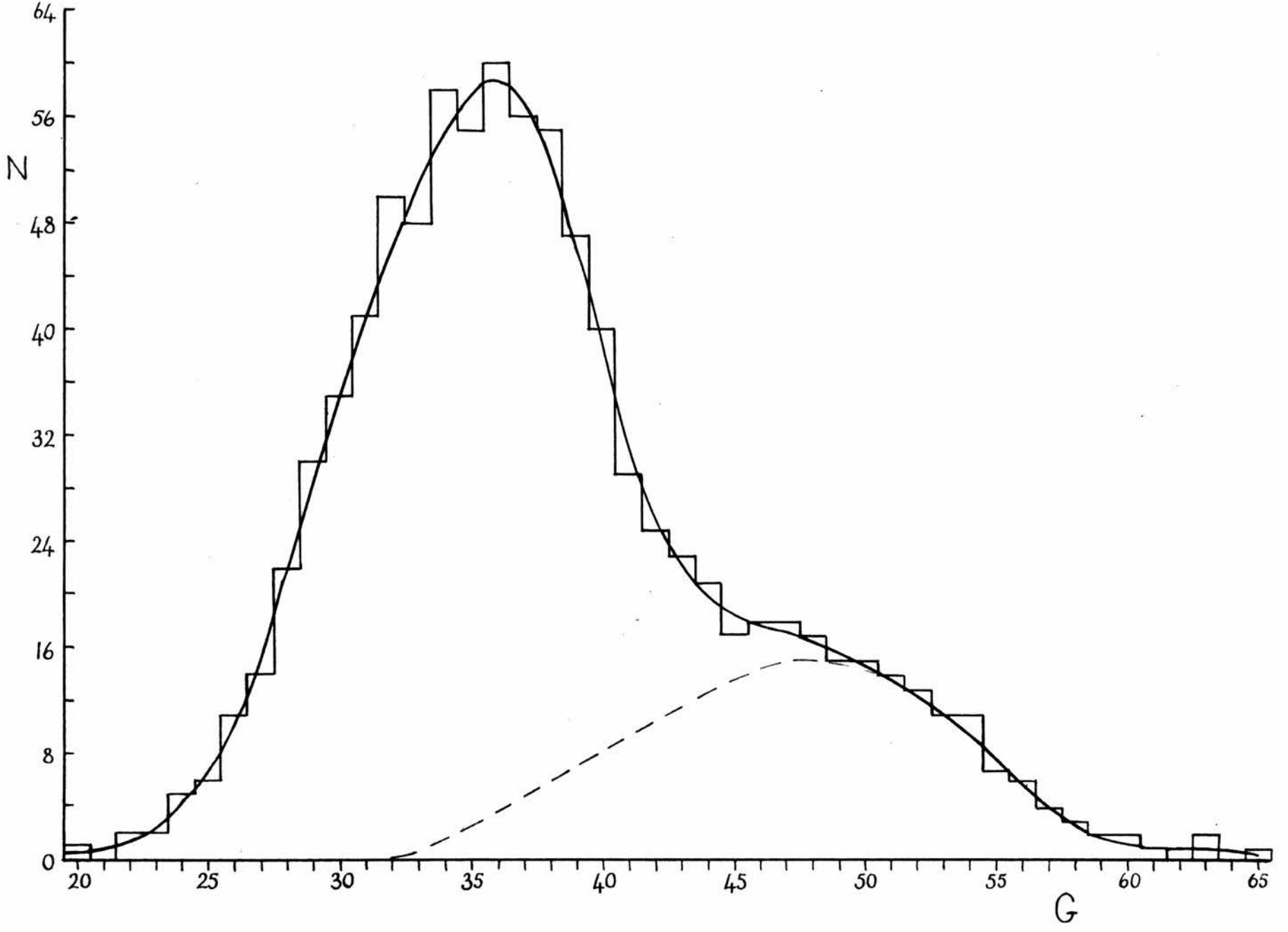

FIGURE 14. FREQUENCY (N) VERSUS GRAIN NUMBER (G) OF CONVERSION ELECTRONS EMITTED FOLLOWING THE ^{228}Th DISINTEGRATION IN THE DE-EXCITATION OF THE 84 keV LEVEL OF ^{224}Ra (PLATE 1).

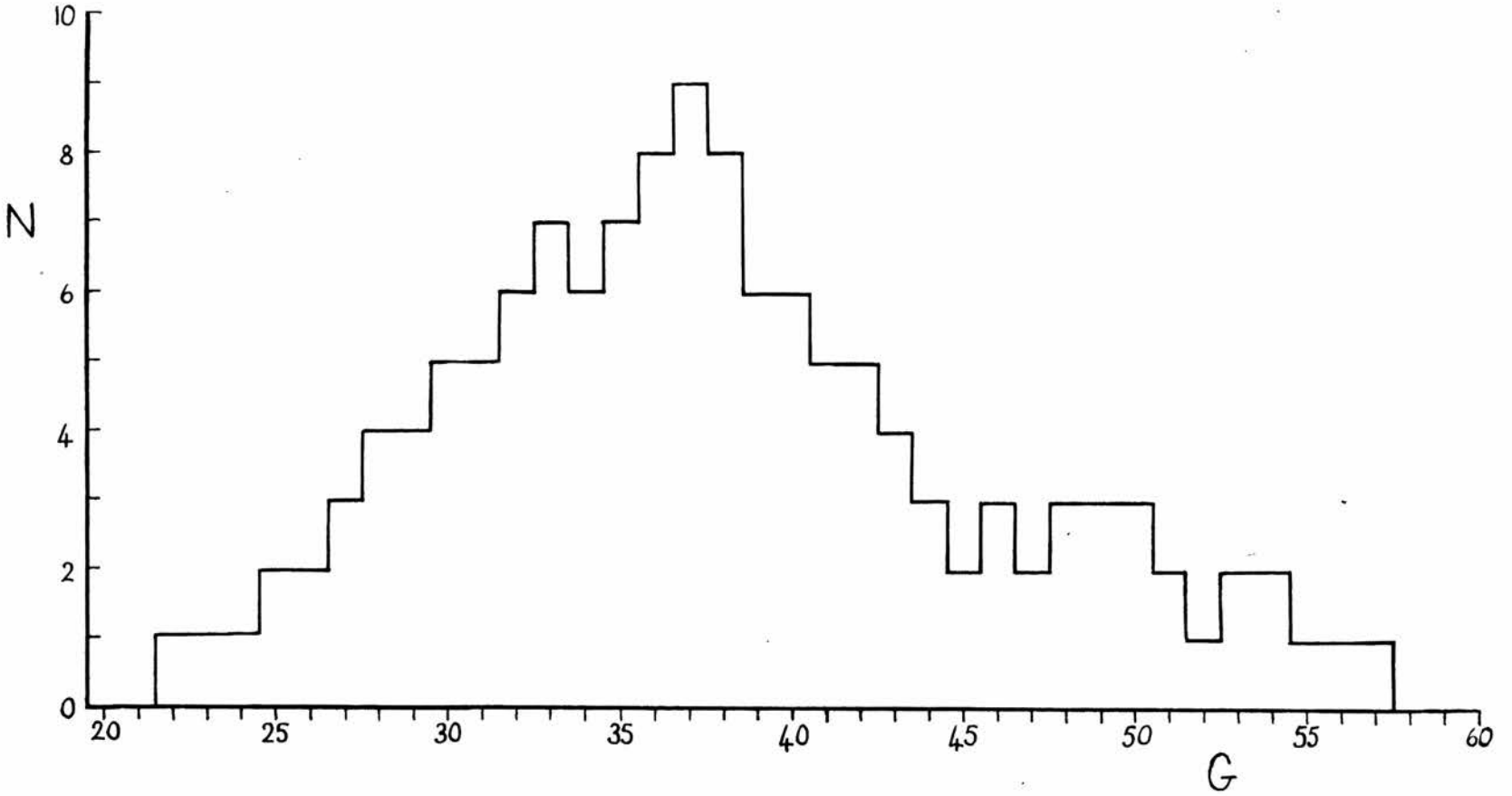

FIGURE 15. FREQUENCY (N) VERSUS GRAIN NUMBER (G) OF CONVERSION ELECTRONS EMITTED FOLLOWING THE ^{228}Th DISINTEGRATION IN THE DE-EXCITATION OF THE 84 keV LEVEL OF ^{224}Ra (PLATE 2).

electron which had been emitted in the de-excitation of a higher excited level, although, as before, a count was kept of the number of all conversion electrons.

The groups of conversion electrons found in Plates 1 and 2 are shown in Table 3.

V.2. Electron grain number versus energy calibration.

Figure 14 shows a histogram and smoothed curve of the grain numbers of 913 conversion electrons associated with the disintegration of ^{228}Th and interpreted as having been emitted in the de-excitation of the 84 keV level of ^{224}Ra. This distribution indicates the existence of two groups with peak grain numbers of 36 and 47. The distribution was resolved into its component groups, by the same method as described in Chapter IV. The half-widths at half, quarter and three quarters of the peak were calculated from the low energy side of the main group as 18%, 25% and 11% of the peak grain number respectively. With these percentages and the ratio of the intensity of conversion in the L-shell to that of conversion in the M- and higher shells (Rosenblum, Valadares and Guillot 1952; 1954) the most probable grain number of the component groups were found to be $35\frac{1}{2}$ and $47\frac{1}{2}$. The standard deviations of both component distributions

Table 3 continued.

Total 9,850 events

	Mean Grain Number	Number of Events	Intensity
Group 1	$35\frac{1}{2}$, $47\frac{1}{2}$	2650	27 ± 1%
Group 2	13	4(2)	0.04 ± 0.02%
Group 3	70	1	-
Group 4	30 or 48	1(1)	-
Group 5	8	8(3)	0.08 ± 0.03%
Group 6	14	4(1)	0.04 ± 0.02%

The number of doubtful events in each group is shown in brackets. These events are not taken into consideration in the calculation of the intensities. The electrons in groups 2 and 4, and one electron in group 5 are associated with a second electron which was interpreted as having been emitted in the de-excitation of the 84 keV level.

were calculated as 13% of the peak grain numbers.

Accurate values have been given, by the above authors (1954), for the energies of the various groups of conversion electrons emitted in the de-excitation of the 84 keV level of ^{224}Ra. From the quoted energies and relative intensities of these groups, the energies expected for the L- and M + higher shells, as unresolved groups, were estimated to be 67.5 keV and 81.0 keV respectively. By assigning the grain numbers of $35\frac{1}{2}$ and $47\frac{1}{2}$ to these energies, two points were obtained for the grain number versus energy calibration. A third point was obtained by using the mean grain number of the electrons in group 5. This group contained eight electrons with a mean grain number of 8; the mean range of these electrons is $3.65 \pm 0.05\mu$. From the mean range versus energy relation of Zajac and Ross (Figure 6) the associated energy was estimated as 20.0 ± 0.2 keV. With this energy and the mean grain number of 8, the third point was obtained for the grain number versus energy calibration (see Figure 16).

Before the experimental results are interpreted, the level scheme suggested by Asaro, Stephens and Perlman will be discussed.

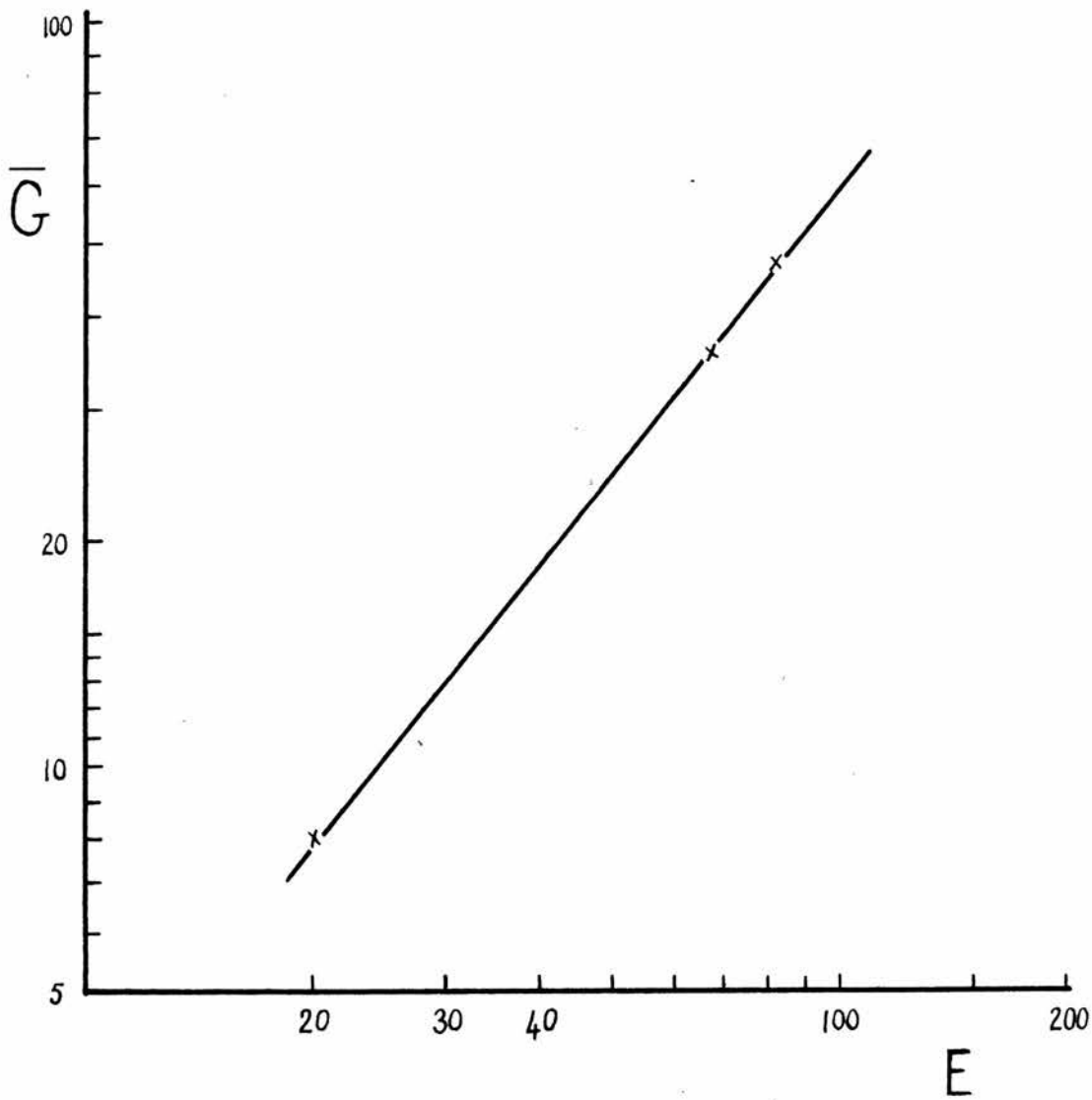

FIGURE 16. MEAN GRAIN NUMBER ($\overline{G}$) VERSUS ENERGY IN keV (E) FOR ELECTRON TRACKS - CALIBRATION.

V.3. Discussion of the level scheme shown in Figure 3.

The scheme suggested by Asaro, Stephens and Perlman shows levels with spins and parities of 0 +, 2 +, 1 - and 4 +. The even parity levels are in agreement with the predictions of the Bohr-Mottelson collective excitation theory (1953).

This theory predicts levels of a rotational character in the even-even nuclei, which are far removed from the closed shells, with energies E, spins I and even parity given by

$$E = \frac{\hbar^2}{2J} I(I + 1)$$

where I = 0, 2, 4 .... and J is the effective moment of inertia. These levels have the quantum number K equal to zero, where K is the component of I along the intrinsic symmetry axis.

The first excited level of ^{224}Ra is known to have spin 2 and even parity, so that if the above rule is followed exactly it is to be expected that the 4 + level would have an energy of 282 keV. In fact, deviations from this simple rule occur as the closed shell, of 82 protons and 126 neutrons, is approached, in such a way as to depress the higher energy levels proportionately more than the lower ones.

In a ~~confined region,~~ narrow range of A and Z a low-lying excited level appears, which is not a member of this rotational sequence. This level apparently has a spectroscopic

designation of 1 - and was first recognised in the ^{228}Th disintegration. There are now six cases, ^{222}Ra, ^{224}Ra, ^{226}Ra, ^{226}Th, ^{228}Th and ^{238}Pu in which this level has been found, lying either between the 4 + and 2 + levels or just above the 4 + level. The type of configuration for an odd-parity level, at an energy of a few hundred keV above the ground state, has not yet been decided. However, a suggestion has been made that such a state may have the same intrinsic structure as the ground state and represent a collective distortion in which the nucleus is pear-shaped. If this were the case, then the 1 - state would have the quantum number $K = 0$, a supposition which can be checked by comparing the γ-transition probabilities to the 0 + and 2 + levels.

By following the treatment of Alaga, Alder, Bohr and Mottelson (1955), it is possible to calculate the theoretical ratio of the reduced transition probability for the γ-radiation between the 1 - and 0 + levels to that for the γ-radiation between the 1 - and 2 + levels. This ratio can take the value of 0.5 or 2.0 depending on whether the 1 - level is a $K = 0$ or $K = 1$ state. Stephens, Asaro and Perlman (1955) obtained an experimental value for this ratio by removing the expected third-power Weisskopf energy dependence from the observed γ-ray intensities. For

the above mentioned isotopes, the values varied from 0.36 ± 0.15 (^{224}Ra) to 0.60 ± 0.15 (^{238}Pu) and these workers therefore concluded that the 1 - levels are K = 0 states.

One other point is worth mentioning at this stage. In ^{228}Th, which results from the β - disintegration of ^{228}Ac, there exists a 3-level at 395 keV above the ground state (Nielson*). This level is separated by 67 keV from the 1 - level and, since the energy spacing between the 0 + and 2 + states is 58 keV, its existence suggests that a second rotational sequence is appearing. With this interpretation, the 3-level would be a K = 0 state.

V.4. Interpretation of the Experimental Results.

The conversion coefficients used in the following considerations were calculated from data given by Rose, Goertzel and Swift (private communication).

V.4.1. Group 1.

As stated above, this group of conversion electrons was interpreted as having been emitted in the de-excitation of the 84 keV level of ^{224}Ra. These electrons were emitted with an intensity of 27 ± 1% of the ^{228}Th α-emission. The results obtained by

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* A paper presented at a conference of the Physical Society in December, 1956.

Rosenblum, Valadares and Guillot (1952; 1954) indicate that 74% of these conversion electrons were emitted from the L-shell and the remainder from the higher electron shells. This leads to an intensity of 20 ± 1% for the L-conversion electrons. The L-shell conversion coefficient for E2 radiation of 84 keV was estimated as 19, leading to a γ-ray intensity of 1.05 ± 0.05%. The sum of the electron and γ-ray intensities gives an intensity of excitation of the 84 keV level in good agreement with that of 28% found from the α-particle spectrum.

This γ-ray intensity is lower than the values given by Asaro (1.6%) and by Stephens (1.7%). There are two possible explanations of this discrepancy (i) Asaro and Stephens, as well as detecting the γ-radiation from the 84 keV level, also detected the K X-radiation of radium, which has an energy of about 86 keV. (ii) the conversion coefficient, used in the present consideration, is too high. The first of these explanations is ruled out by the fact that Newton and Rose (1954) placed an upper limit, of 5% of the 84 keV radiation, on the intensity of the K X-radiation emitted following the disintegration of ^{228}Th. The second explanation is not ruled out so easily since it is possible that the estimated conversion coefficient of 19 is too high. In order to obtain a γ-ray

intensity of 1.6% or 1.7%, it is necessary to postulate a conversion coefficient of about 12, which is consistent with the values suggested by previous workers.

One point should be noted at this stage. Some of the electrons of this group will have been associated with the de-excitations of the higher excited levels in ^{224}Ra. These levels are de-excited by the emission of γ-rays and conversion electrons and some of the transitions lead to the 84 keV level which, in turn, is de-excited mainly by the emission of conversion electrons. In the cases in which the first transition occurred by means of conversion electrons, the existence of two electron tracks associated with one α-particle track indicated the existence of the cascade process. If, however, the initial transition occurred by γ-radiation, only one electron track would have been produced and such events would be interpreted as belonging to the α-disintegration to the 84 keV level. Using the values given by Asaro, Stephens and Perlman for the intensities of the γ-ray feeds to the 84 keV level, it was estimated that approximately 35 electrons from this group should be attributed to the de-excitations of the higher energy levels. The intensity of the conversion electrons associated with α-particles feeding the first excited level is reduced but not by an amount sufficient to

alter the value of 27 ± 1% given above.

Mean Range of the associated α-particles.

Figure 17a shows the frequency distribution of the ranges of 1,500 α-particles. Of these, 42 were attributed to the disintegration of ^{224}Ra and, thus, 1,458 were considered to have been emitted from ^{228}Th. This α-particle group consists effectively of two unresolved sub-groups, one feeding the ground state of ^{224}Ra and the other feeding the first excited level. The mean range, with standard error, of the 1,458 α-particles is 23.43 ± 0.02 μ and represents the mean range in G5 emulsions of the two groups of α-particles of energies 5.421 MeV and 5.338 MeV. It is known, however, that the α-particles associated with the conversion electron, which had been emitted in the de-excitation of the 84 keV level of ^{224}Ra, must belong to the lower energy group. The mean range of 927 such α-particles is 23.12 ± 0.02 μ (Figure 17b). In order to determine whether or not the difference between these mean ranges, 0.31 ± 0.03 μ, could have resulted from purely random sampling, a Student's "t-test" was applied. The value of t was calculated to be 10.13 and, since t = 2.58 indicates a probability of obtaining such a difference by chance of 1 in 100, the two means can be taken as significantly different and to represent two α-populations. The value of

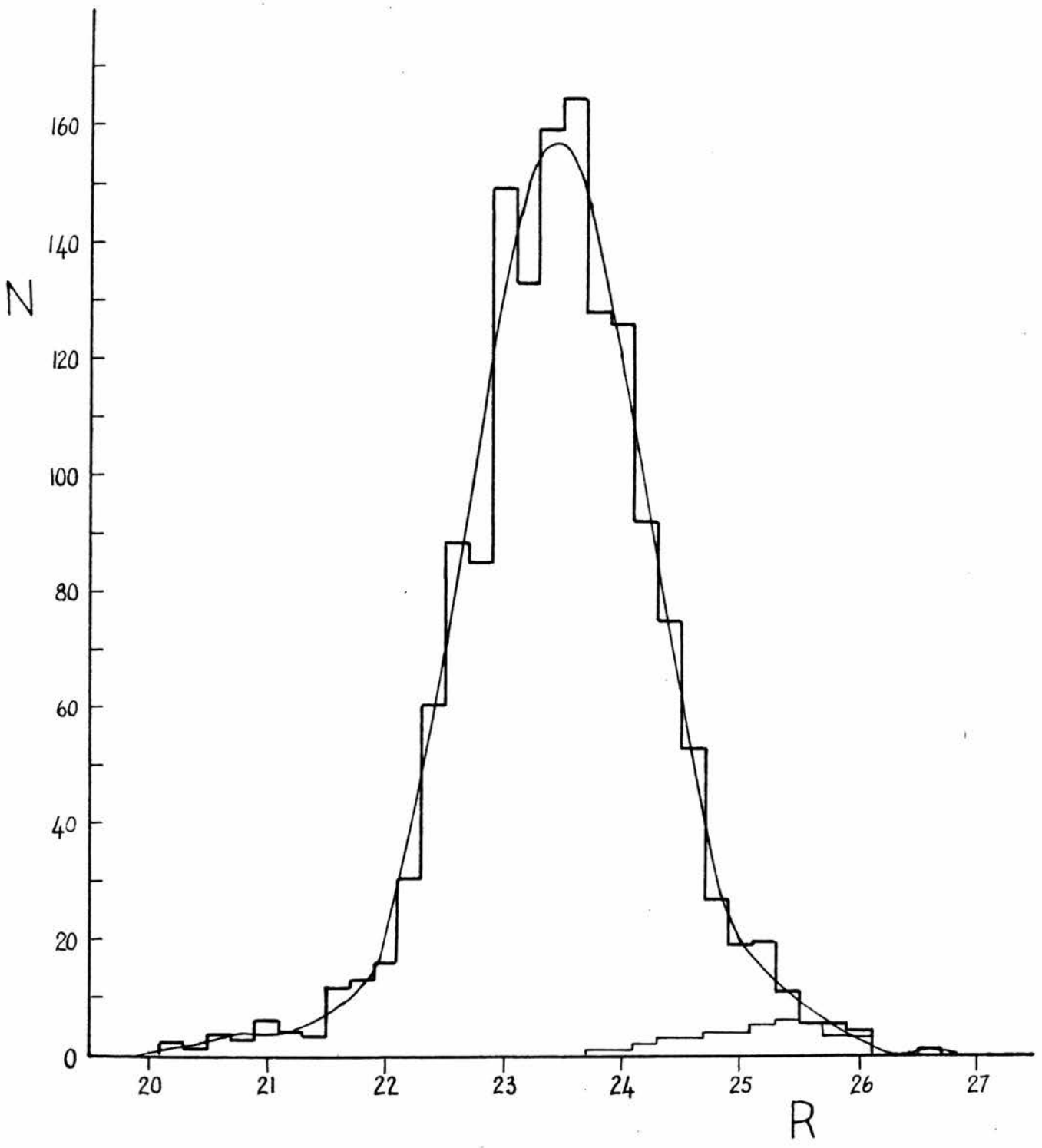


FIGURE 17a. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF "ALL" ^{228}Th α-PARTICLES (PLATE 1).

The half-width at half maximum is 0.9 μ. The distribution of the ^{224}Ra α-particle tracks thought to be present is shown under the high energy tail.

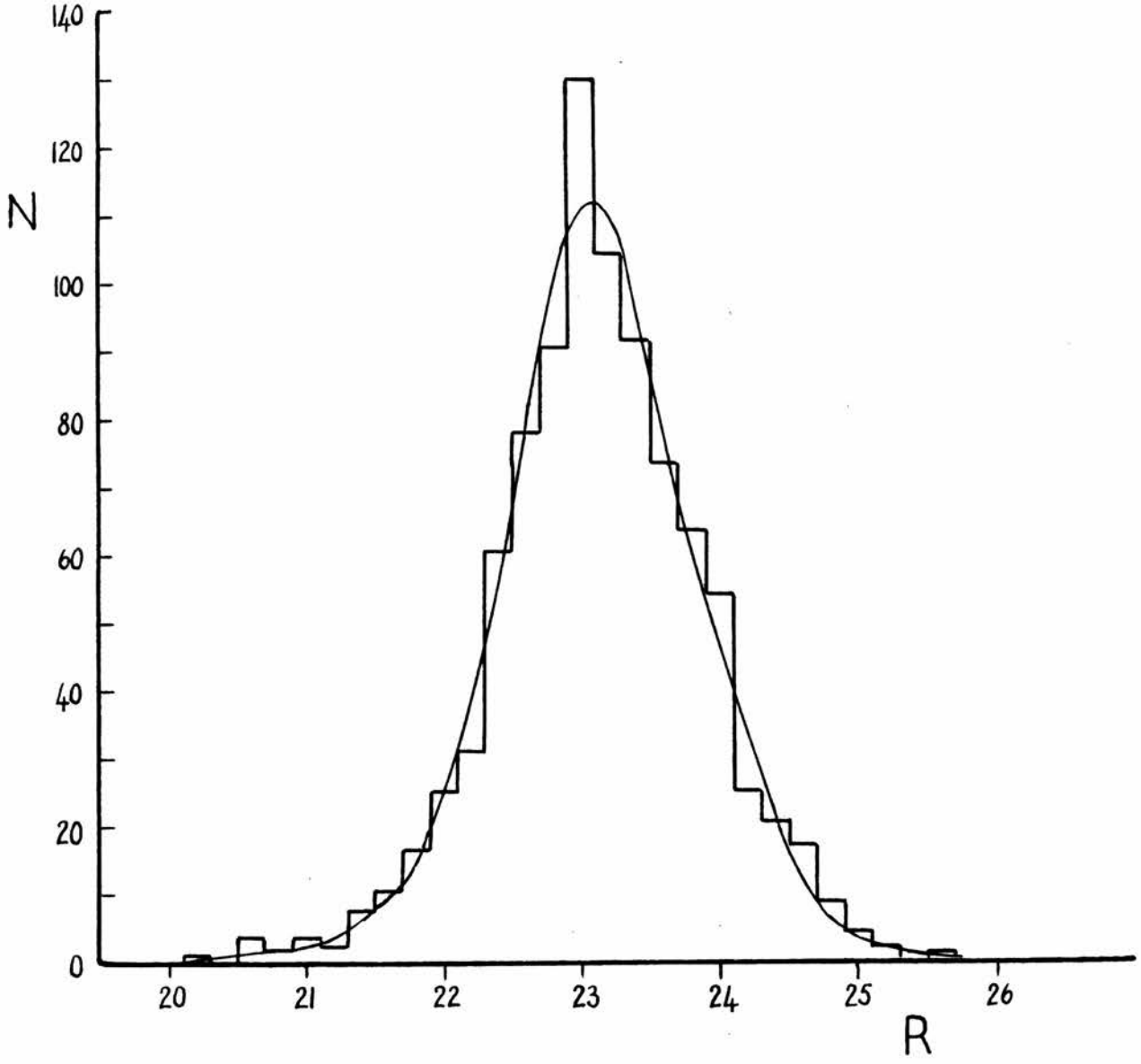


FIGURE 17b. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF ^{228}Th α-PARTICLES WITH ASSOCIATED CONVERSION ELECTRONS (PLATE 1).

The half-width at half maximum is 0.75 μ.

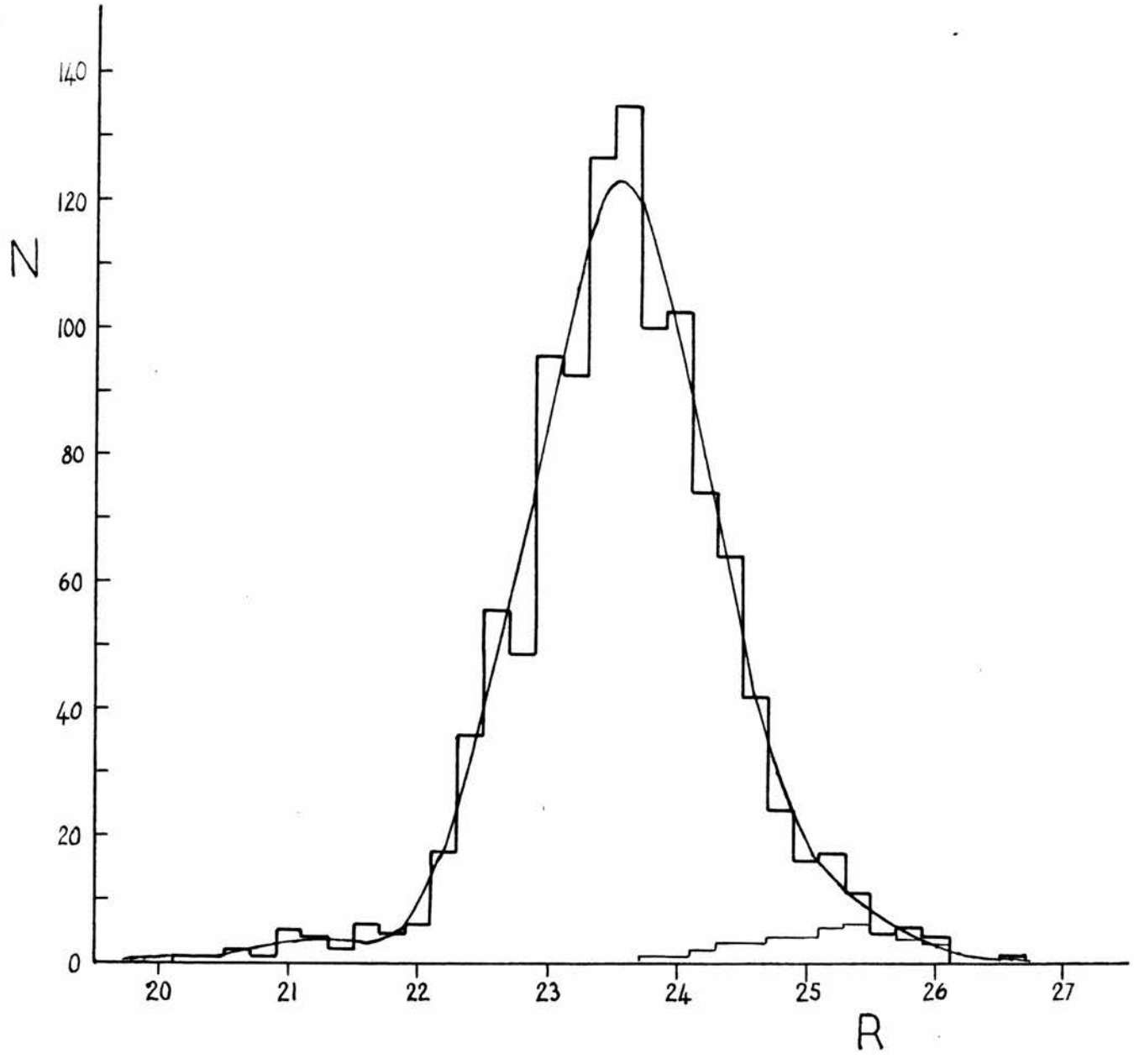


FIGURE 17c. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF ^{228}Th α-PARTICLES WITH NO ASSOCIATED CONVERSION ELECTRONS (PLATE 1). The half-width at half maximum is 0.80μ. The distribution of the ^{224}Ra α-particle tracks thought to be present is shown under the high energy tail.

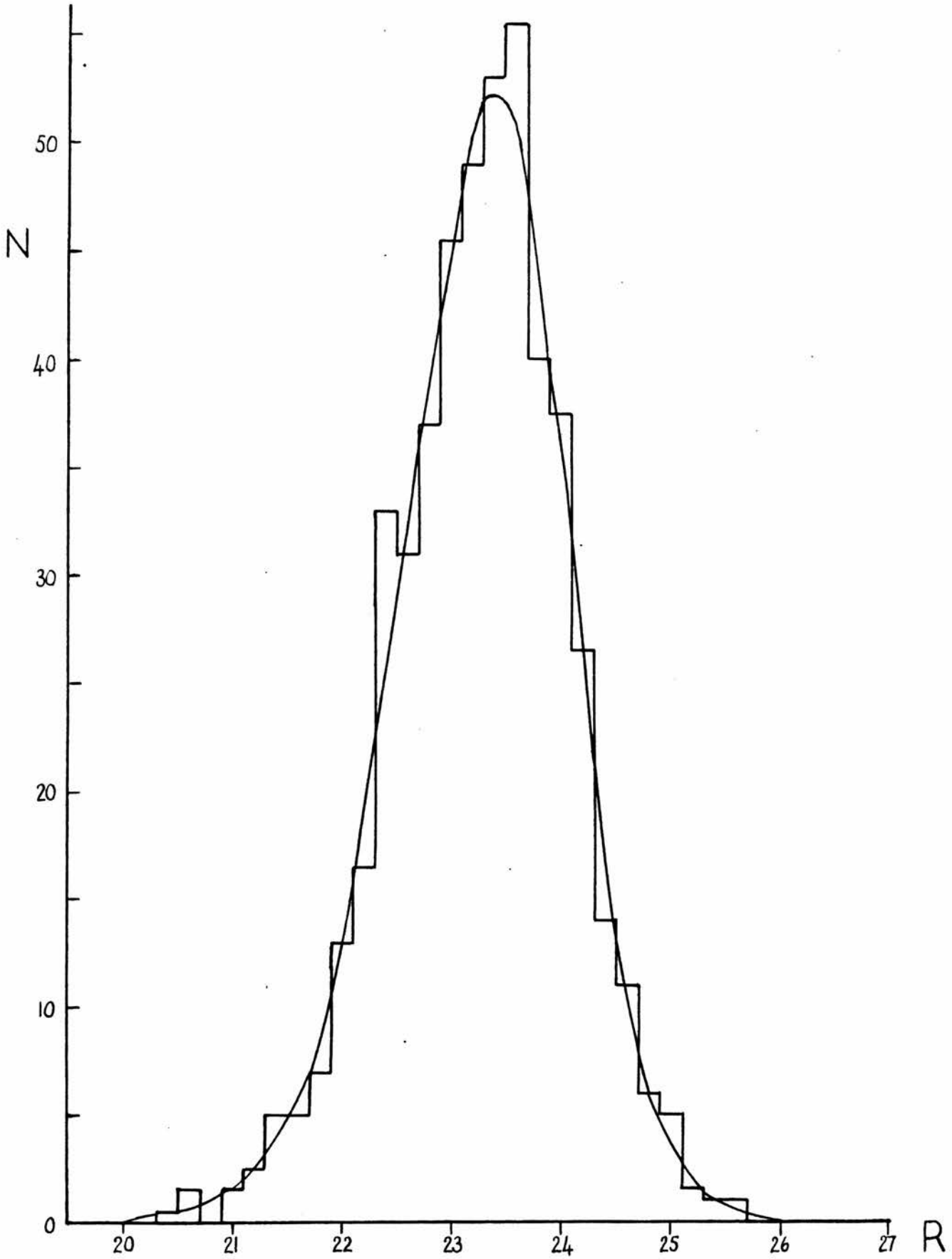


FIGURE 18. FREQUENCY (N) VERSUS RANGE IN MICRONS (R) OF "ALL" ^{228}Th α-PARTICLES (PLATE 2).

The half-width at half maximum is 0.9 μ .

23.12 ± 0.02 μ is, therefore, considered to be the mean range of α-particles of energy 5.338 MeV.

In order to examine if the range of the α-particles emitted in the normal disintegration to the ground state of ^{224}Ra is significantly greater than the mean range of all α-particles, an estimate was made of the mean range of 1,064 α-particles which were not associated with electrons. The mean range, with standard error, of these tracks was found to be 23.55 ± 0.03 μ. The calculated t-value of 3.53 indicates that the difference between the means of 0.12 ± 0.04 μ is significant and the mean G5 emulsion range of energy 5.421 MeV is therefore taken as 23.55 ± 0.03μ.

The difference of 0.43 ± 0.04 μ between the mean ranges of the α-particles feeding the ground state and first excited level of ^{224}Ra represents a difference of 90 ± 8 keV between the kinetic energies of the two α-particle groups. This energy is consistent with the energy of 84 keV attributed to the first excited level.

The above consideration is limited to the α-particle ranges measured in Plate 1. The mean range of 500 α-particles, observed in Plate 2, is 23.41 ± 0.03 μ which agreed with the corresponding value obtained from Plate 1.

V.4.2. Group 2.

The electrons of this group, with a mean grain

number of 13 and a standard deviation of ± 1 grain, are associated with an energy of 30 ± 2 keV; the energy was estimated from the calibration curve shown in Figure 16. These electrons were interpreted as having been emitted in the de-excitation of a higher excited level since in each case a second electron track was observed. The grain numbers of these longer tracks were consistent with that expected for L-conversion electrons emitted in the 84 keV transition and indicated that a cascade process, involving the first excited level, had occurred. The binding energies of electrons in the K-, L- and M-shells of radium are 104.5 keV, 19.2 - 15.4 keV and 4.8 - 3.10 keV respectively (Cauchois 1952). The conversion electrons could therefore have been emitted in a transition of approximately 134 keV, 47 keV or 34 keV. On the basis of the level scheme shown in Figure 3, two alternative interpretations are possible; the electrons could have been emitted from the K-shell in a transition of 134 keV occurring between the second and first excited levels, or from the M-shell in a transition of 34 keV, which may have occurred between the third and second excited levels, and been followed by a γ-ray transition to the first excited level. The following fact favours the former interpretation. If the electrons had been emitted from the M-shell, then it is to be expected

that some electrons would have been emitted from the L-shell. No tracks, associated with a second electron track and which could be so interpreted, were observed.

The intensity of this electron group is 0.04 ± 0.02%. The intensity of the γ-rays emitted in the 137 keV transition was given by Asaro as 0.26% and, therefore, to reconcile the electron intensity to this, requires that the conversion coefficient is < 1. This excludes the possibility of the radiation being magnetic, but the K-conversion coefficients for E1 and E2 radiation both satisfy the condition. The estimated conversion coefficients for E1 and E2 radiation are given below.

	E1	E2
K-conversion coefficient	0.2	0.3
L-conversion coefficient	0.03	1.2

If the transition were E2 then, from the values of the K- and L-conversion coefficients, it is to be expected that about twelve tracks of electrons emitted from the L-shell should have been observed. While it is possible that some of these tracks could have been missed, their complete absence indicates that the radiation is most probably E1. This is consistent with the multipolarity assigned to the radiation by Asaro, Stephens and Perlman. With the conversion coefficient of 0.2, the γ-ray intensity was estimated as 0.2 ± 0.1% of the

total α-emission.

The mean range of the associated α-particles is $22.6 \pm 0.2\,\mu$, which is lower than the mean range of the α-particles feeding the ground state by $1.0 \pm 0.2\mu$. Although this value cannot be considered significant, it does permit an estimate to be made regarding the difference in the kinetic energies of the two α-particle groups. The difference was estimated as 170 ± 30 keV.

It is therefore concluded that the electrons in this group were emitted in an E1 transition between the second and first excited levels of ^{224}Ra.

V.4.3. Group 3.

Since the electrons, with grain numbers between 72 and 84, found in Plate 1 were probably emitted following the disintegration of ^{224}Ra, only the electrons observed in Plate 2 will be considered in this discussion. The grain number of this track was estimated to be 70 with a standard deviation of 8 grains and, while it is not possible to assign an accurate energy value to this event, the associated energy probably lies in the region between 105 keV and 125 keV. Since the event in which this electron occurred showed no evidence of a cascade process, it was assumed that the transition led directly to the ground state of ^{224}Ra. The most likely interpretation is that the

electron had been emitted from the K-shell in the transition between the second excited level and the ground state. The binding energy of electrons in the K-shell is 104.5 keV and this leads to a nuclear energy difference of between 210 keV and 230 keV, which is consistent with the level scheme shown in Figure 1. The α- γ angular correlation measurements (Stephens) indicate that the radiation is E1 but, since only one event has been observed in the present investigation, no attempt was made to estimate the intensity of the γ-radiation. For the same reason, no reliance can be placed upon the range of the associated α-particle.

V.4.4. Group 4.

The only event observed, which fell into this group, consisted of two electron tracks associated with the one α-particle track and indicated a cascade process. The grain numbers of the electron tracks were estimated as 30 and 48 and the most likely interpretation is that they were emitted in the de-excitation of the third excited level. It is not possible to say definitely which electron belonged to the 84 keV transition, since a grain number of 30 is consistent with that expected for an electron emitted from the L-shell and that of 48 is consistent with that expected for an electron emitted from a higher shell (see Figure 14).

On the first assumption the 48 grain electron,

with an estimated energy in the region between 75 keV and 95 keV, could be interpreted as having been emitted from the K-shell in a transition of 180 keV to 200 keV whereas, on the second assumption, the 30 grain electron, with an estimated energy between 50 keV and 65 keV, could be interpreted as having been emitted from the K-shell in a transition of 155 keV to 170 keV. Since the energy estimated from the grain number of a single electron track cannot be considered accurate, it is not possible to decide which interpretation is correct but both are consistent with the view that the electrons were emitted in the de-excitation of the third excited level.

No attempt was made to assign a polarity to the initial transition in the cascade process. If, as has been suggested by Asaro, Stephens and Perlman, the 169 keV radiation is E2, then the ratio of the K-conversion coefficient to the L-conversion coefficient (approximately 1:3) indicates that a few L-conversion electrons should have been emitted. No events, which could be so interpreted, were observed. This, by itself, cannot be taken as conclusive evidence that the radiation is not E2, since the tracks produced by such electrons, with initial energies of about 150 keV, would have low grain densities at the high energy end and, if unfavourably presented in the field of view, could have been missed. If, on the other hand, the

radiation were E1, no L-conversion electrons would be expected. No significance was attached to the range of the associated α-particle.

So far, the results obtained from this investigation may be considered consistent with the de-excitation scheme shown in Figure 3.

V.4.5. Groups 5 and 6.

The electrons in these groups were interpreted as having been emitted in the same transition, giving a total intensity of 0.12 ± 0.04%. Group 5 consists of eight electrons with a mean grain number of 8 ± 1 and has an associated energy of 20 ± 2 keV. Group 6 contains four electrons with a mean grain number of 14 ± 1 and an associated energy of 32 ± 2 keV. It is possible that these electrons represent only one group, with grain numbers between 6 and 16, but the absence of grain numbers 10 to 12 makes this improbable. If this were the case, the estimated energy associated with the group would be 24 ± 5 keV. The electrons could have been emitted from the K-shell in a transition of 128 ± 5 keV which had occurred between the second and first excited levels. However, Newton and Rose (1954) set an upper limit on the K X-radiation of 0.08% of the total α-emission. The intensity of vacancies, produced in the K-shell by the electrons of groups 2 - 4,

is about 0.06% and, since the fluorescent yields from the K-shells of the heavy elements are very high, it is most unlikely that the electrons at present under consideration were emitted from this shell. If the electrons in this composite group had been emitted from the L-shell in a transition of 41 ± 5 keV some M-conversion electrons should have been observed but if they had been emitted from the M-shell in a transition of 28 ± 5 keV the associated L-conversion electrons, with an energy of 11 ± 5 keV, could have been missed. The most probable interpretation of the two groups of electrons is that they were emitted from the L- and M-shells in a transition of 37 ± 2 keV.

The mean range of the associated α-particles, 21.8 ± 0.2 μ , is lower than that of the α-particles feeding the ground state of ^{224}Ra by 1.8 ± 0.2 μ and this difference corresponds to a difference of 250 ± 30 keV between the kinetic energies of the α-particle groups. This suggests that the electrons were emitted in the de-excitation of a level at least 200 keV above the ground state.

The results obtained, by earlier experiments, from studies of the γ-ray spectrum show no evidence for the existence of a 37 keV transition. This could be due to the unfavourable position in the energy spectrum at which such a transition would occur and to

the low intensity of the γ-radiation, especially if the multipolarity were E2. The present author cannot find any contamination present in the impregnating solution which could give rise to the events observed. It must therefore be considered that these groups represent genuine conversion electrons emitted following the α-disintegration of ^{228}Th. The total intensity of the transition could be either 0.21 ± 0.07% if the radiation were E1 (conversion coefficient = 0.86) or 0.12 ± 0.04% if the radiation were E2 (conversion coefficient ≈ 700).

The following discussion is an attempt to find a level scheme, consistent with the results obtained from the investigations of the α-particle and γ-ray spectra, into which the 37 keV transition can be fitted.

V.5. Consideration of possible level schemes.

The results obtained from studies of the ^{228}Th α-particle spectrum show the existence of two levels in ^{224}Ra which lie more than 200 keV above the ground state, one at 217 keV and the other at 253 keV.

If the electrons were interpreted as having been emitted in a transition de-exciting the 217 keV level, it would be necessary to postulate the existence of an intermediate level, either 180 keV or 37 keV above the ground state, depending on whether the electrons under

consideration fed or de-excited this level. It is already known that the 217 keV is de-excited by the emission of two γ-rays with E1 multipolarity and, therefore, if a third mode is to compete favourably with these it would have to be E1, or possibly E2. Such a transition would then require that the intermediate level, whether at 180 keV or 37 keV, had a spectroscopic designation of 0 + or 2 +, if the radiation were E1, or 1 - or 3 - if the radiation were E2. An excited level with any of these spin and parity values would be fed by α-particles and no α-lines, which could be so interpreted, have been observed. No γ-radiation of 180 keV has been observed in the γ-ray spectrum. In the above consideration, it is assumed that the de-excitation process involves only two transitions but more elaborate schemes are also ruled out for similar reasons.

On the other hand, if these electrons are assumed to be involved in the de-excitation of the 253 keV level and if they are emitted in the first part of the cascade process, the intermediate level would lie at 216 keV above the ground state, which is consistent with the known level at 217 keV. This level would be de-excited, in turn, by the transition discussed in V.4.2 and V.4.3. The possible interpretation, that the 216 keV γ-ray was emitted before the electrons, can be

ruled out for reasons similar to the above.

If this explanation is correct, then the 253 keV level cannot be a 4 + state. This level would be de-excited by two competing transitions, one of 169 keV, leading to the 2 + level and the other of 37 keV, leading to the 1 - level. If, as has been suggested, the 169 keV radiation is E2, then the 37 keV would be E1. Such transitions lead to the assignation of 0 + to the 253 keV level. With the multipolarities reversed, the spectroscopic designation would be 3 - (see Figures 19a, b).

These level schemes and other possibilities will now be discussed in detail. In Figure 19 the various level schemes are shown; the established transitions are indicated by the heavy lines and the suggested transitions by the broken lines.

Figure 19g.

This level scheme would necessitate the existence of a fifth line in the α-particle spectrum leading to a level 290 keV above the ground state and no such line has been observed. A more serious drawback to this scheme is revealed by a consideration of the γ-transitions which would de-excite the 290 keV level. If, as is suggested, this level has a spin of 3 and odd parity, it would be de-excited by three transitions, which may or may not compete favourably with one

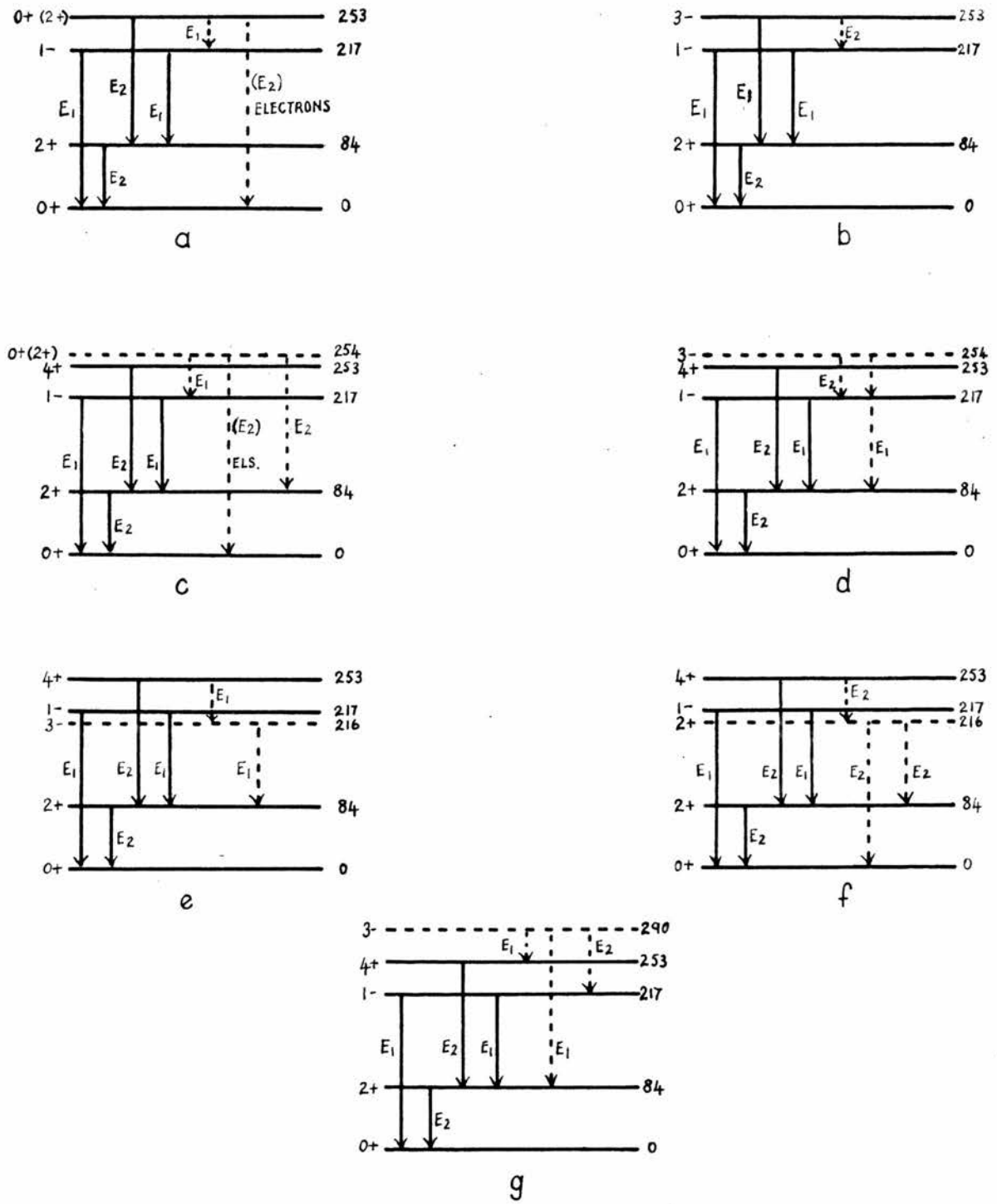


FIGURE 19. SOME SUGGESTED LEVEL SCHEMES OF ^{224}Ra.

See discussion in the text.

another (i) the 37 keV transition to the 4 + level, (ii) a 206 keV transition to the 2 + level and (iii) a 73 keV transition to the 1 - level. The multipolarities of these transitions would be E1, E1 and E2 respectively.

By following the treatment suggested by Alaga, Alder, Bohr and Mottelson (1955), the expected ratio of the transition probabilities for the 206 keV and 37 keV E1 radiations can be calculated. In general, the transition probability per second for γ-emission of frequency ω and multipole order L is given by

$$T = \frac{8\pi(L+1)}{L[2(L+1)!!]^2} \frac{1}{\hbar} \left(\frac{\omega}{c}\right)^{2L+1} B(L) \text{ ---------- (1).}$$

B(L) is the reduced transition probability which can be expressed as a product of the geometrical factor, depending only on the angular momenta I, K, L, and a factor involving integration over the intrinsic wave function of the initial and final states. However, when the reduced transition probabilities for the emission of a given multipole radiation from a state, i, to different members, f, f', of a rotational family are compared, the factors involving the intrinsic wave function are the same and the ratio depends only on the geometrical factors.

$$\frac{B(L\, I_i \longrightarrow I_f)}{B(L\, I_i \longrightarrow I_{f'})} = \frac{\langle I_i\, L\, K_i\, K_f - K_i \mid I_i\, L\, I_f\, K_f \rangle^2}{\langle I_i\, L\, K_i\, K_f - K_i \mid I_i\, L\, I_{f'}\, K_f \rangle^2} \text{ ---- (2).}$$

The quantities on the right are Clebsch-Gordon coefficients for the addition of angular momenta. In the present case, the γ-transitions are E1, the initial state has a spin of 3 and the final states have spins of 4 and 2. With $K_i = K_f = 0$ for both transitions and by using the expressions for the coefficients given by Condon and Shortley (1953) the ratio of $\frac{T_{206}}{T_{37}} \approx 300$ was found from (1).

Since the 3 - level, if it exists, can be populated by α-particles with an intensity of not greater than 1% of the total ^{228}Th α-emission, this ratio indicates that the 37 keV transition could not be observed. This level scheme is therefore rejected.

The schemes shown in Figures 19c, d, e, f are variations of those shown in Figures 19a, b. The variations can hardly be justified on the basis of the available energy determinations but they have the merit that they permit the retention of the 4 + level. These schemes do not require the existence of an "unobserved" line in the α-particle spectrum since the hypothetical level would be populated by the "α-feed" to one or other of the known levels.

In the consideration of these schemes it is not possible to use the above method. Owing to the lack of information regarding the intrinsic wave functions of the various states, the reduced transition probabilities cannot be calculated.

Figure 19e.

It is possible that the E1 and E2 transitions de-exciting the 4 + level could compete favourably with one another since the E2 radiation is enhanced by the available energy and by the fact that it occurs between members of a rotational family. The 3 - level would be fed, by the 37 keV transition, with an intensity of 0.21 ± 0.07%. This level would then be de-excited by an E1 transition of 132 keV, leading to the 2 + level. The γ-radiation from this transition would be part of the observed radiation between the 1 - and 2 + levels. With this interpretation, an intensity of 0.09 ± 0.05% would belong to the de-excitation of the 1 - level. Such an intensity would not necessarily invalidate the conclusion of Stephens, Asaro and Perlman that the 1 - level has the K quantum number equal to zero but it would increase the experimental value which they calculated for the ratio of the reduced transition probabilities.

In all probability the 3 - level would be fed by α-particles. While an accurate estimate of the intensity of this feed cannot be made, it is to be expected that the intensity would be less than that of the α-feed to the 1 - level owing to the difference in the spins. This effect would result in an increase in the intensity of the 132 keV radiation and a

corresponding decrease in that of the 133 keV radiation. The intensity of the 217 keV radiation would also be reduced but the overall result of the α-feed to the 3 - level would be a further increase in the experimental value of the ratio calculated by the above investigators which might be large enough to destroy the basis on which K = 0 was assigned to the 1 - level. This level scheme is therefore suspect.

Figure 19f.

When both the 37 keV and 169 keV transitions are E2, it is most unlikely that the 37 keV transition could be observed. This scheme can therefore be rejected without further consideration.

Figure 19c.

On the basis of the multipolarity assigned to the 37 keV transition, the hypothetical level could be either a 2 + or a 0 + state. If this level were a 2+ state, the 254 keV E2 radiation, which would occur between this level and the ground state, should be observed. Since no γ-rays with this energy have been found, the spin assignment of 2 is doubtful.

If, on the other hand, the hypothetical level were a 0 + state, the modes of de-excitation would be (i) the 37 keV E1 transition to the 1 - level, (ii) the 170 keV E2 transition to the 2 + level and (iii)

conversion electrons, with an energy of at least 150 keV ($0^+ \longrightarrow 0^+$ transition). If the 170 keV transition can compete favourably with the E1 transition, the emitted γ-rays would be observed as part of the 169 keV radiation between the 4^+ and 2^+ levels. If any conversion electrons are emitted, the tracks so produced in the emulsion could be missed owing to their low grain density at the high energy end. This scheme is therefore a possibility.

Figure 19d.

The 3 - level in this scheme would be de-excited by (i) the 37 keV E2 transition and (ii) the 170 keV E1 transition. Unless the E2 radiation were greatly enhanced due to its occurring between two levels which may be members of a rotational sequence, it would not be observed. This scheme therefore appears to be improbable.

Figures 19a and b.

The comments on these schemes are similar to those applied to the schemes of Figures 19c and d respectively.

From these considerations, the most probable schemes are those shown in Figures 19a and c, with the uppermost level having a spin of 0 and even parity,

although the scheme in Figure 19e cannot be ruled out completely.

The formula given by Feather (1952) yields some information regarding the relative intensities of the α-particle feeds to the excited levels.

$$\log_{10}\left(\frac{\lambda_1}{\lambda_r}\right) + 1{\cdot}725\,\frac{A-2}{A}\,(Z-2)\left(\eta_1^{-\frac{1}{2}} - \eta_r^{-\frac{1}{2}}\right) + 2{\cdot}36\left(\frac{A-4}{A}\right)^2 (Z-2)^{-\frac{1}{2}}\,\rho_0^{\frac{3}{2}}\,(\eta_1 - \eta_r) = C\,(Z-2)^{-\frac{1}{2}}\,\rho_0^{-\frac{1}{2}}\,(j_r - j_1)(j_r + j_1 + 1)$$

where the λ s are the partial disintegration constants, the η s are the disintegration energies in MeV, the j s are the values of the spin change, ρ_0 is the nuclear radius in units of 10^{-12} cm., taken as 0.889, in this case and C is a constant equal to 0.71 ± 0.01.

By calculating the ratio $\frac{\lambda_1}{\lambda_r}$ for the disintegrations to the ground state and an excited level of 0 + at 253 keV, the intensity of the α-particle feed to the excited level was found to be 2.4% of the total ^{228}Th α-emission. While the formula is not expected to give accurate values in cases where the spin change is small, this result suggests that the existence of a level 0 + at 253 keV is rather improbable. The calculated intensity for a 3 - level 253 keV above the ground state is 0.25% and that for a 4 + level is

0.06%. Therefore, it is tempting to suppose that the schemes shown in Figures 19b and d are possibilities.

If one of these is the correct scheme, the 37 keV E2 radiation must be greatly enhanced by occurring between members of a rotational sequence. Assuming that this rotational band is governed by a similar equation to that given on page 65, then

$$E_{even} = \frac{\hbar^2}{2J} I(I+1) \qquad I = 0, 2, 4 \ldots \text{ even parity}$$

$$E_{odd} = \frac{\hbar^2}{2J'} I'(I'+1) \qquad I' = 1, 3, \ldots\ldots \text{ odd parity}$$

where E_{even} and E_{odd} are the energies of the levels in the rotational sequences commencing with the 0 + and 1 - states respectively. Since the energy spacing between the 0 + and 2 + levels is 84 keV and that between the 1 - and 3 - levels is approximately 37 keV $\frac{J'}{J} \approx 4$. Thus the onset of this odd parity rotational sequence would require a large increase in the effective moment of inertia.

It is obvious that the problem of deciding upon the correct level scheme cannot be solved with the data which is at present available and that further experiments are necessary.

The electrons of groups 5 and 6, with the exception of one electron in group 5, are not associated with a second conversion electron track. The intensities of the 212 keV and 137 keV γ-rays, as given by Asaro, Stephens and Perlman, indicate that approximately half of the de-excitation of the 217 keV level takes place by way of the 137 keV transition in cascade with the 84 keV transition. If these electrons feed the 217 keV level it is to be expected that, of the 12 events observed, 6 should show an 84 keV transition electron and 1 or 2 should also show the existence of an electron from the 137 keV transition. One electron in group 5 was associated with an electron emitted in the de-excitation of the 84 keV level and another event was observed in which the α-particle was apparently associated with three conversion electron tracks but the interpretation was impossible. It therefore appears that some low energy electron tracks have been missed in the events which showed an 84 keV transition electron track. This is not unreasonable, since the low energy track could be obscured, or misinterpreted as part of the longer electron track. Therefore, the correct composite intensity of these two conversion electron groups is probably higher than the value quoted earlier.

V.6. Suggestion for further experiments.

These experiments would be designed to establish initially two points (i) that the 37 keV transition follows the α-disintegration of ^{228}Th and (ii) the multipolarity of this transition.

If this radiation were E1 the intensities of the L_I, L_{II} and L_{III} conversion electrons would be in the ratio of 0.26:0.29:0.31 (the estimated values of the conversion coefficients) but if it were E2 the ratio would be 10:365:332. Therefore, by comparing the intensities of the three conversion lines, the multipolarity of the transition can be determined. In principle, the following experiment enables this comparison to be made.

A solution containing a few millicuries of ^{228}Th, purified from its disintegration products, would be evaporated to small volume. The residue would be placed on a loop of platinum wire, diameter 0.3 mm., bent into the shape of a hairpin and evaporated to dryness. The wire would then be straightened and clamped into the source holder of a β-spectrometer. The conversion electrons would be detected with an Ilford Selochrome 120 photographic plate which, after an exposure of a few hours, would be processed, examined under a microphotometer and the relative intensities of the L_I, L_{II} and L_{III} conversion lines

estimated. Assuming that the energy of the transition is exactly 37 keV, the energies of the three lines are 17.77 keV, 18.52 keV and 21.56 keV respectively and, therefore, by using a magnetic field of 100 oersteds, the radii of curvature would be approximately 4.5 cm., 4.6 cm. and 5.0 cm. With a narrow slit, 0.5 mm., these lines could be resolved.

In the source used, ^{228}Th must be freed from its disintegration products to 1 part in 10^5. Following the α-disintegration of ^{212}Bi (ThC), a level is excited in ^{208}Tl (ThC") with an energy of 40 keV. This level is de-excited mainly by the emission of conversion electrons which might cause confusion if the energy of the "37 keV" transition is higher than expected. The 40 keV transition occurs with an intensity of 70% of the α-emission, which itself occurs in 34% of the disintegrations of ^{212}Bi (see Figure 1). Therefore, in order that the intensity of this transition be less than 1% of that of the L-conversion electrons from the 37 keV transition (taken as 0.08%), it is necessary to reduce the activity of ^{212}Bi to 1 part in 10^5 of the equilibrium amount. To prevent an appreciable growth of this isotope during the exposure time, ^{224}Ra and ^{212}Pb must also be removed from the ^{228}Th to a similar degree. The chemical separation could be performed by a method similar to that

described on page 22, recycling the various stages three or four times, or by the use of an ion exchange column.

Before performing such an experiment, it would be necessary to calibrate the photographic plate since the density of the blackening produced varies with the energy of the electron under consideration (Mladjenović, 1954). In the region of the energies quoted above, the density falls very rapidly with decreasing energy and, therefore, the relative intensities of the three lines, as shown on the plate, would not reflect the true relative intensities of the electrons emitted. Thus, the density versus electron energy curve would have to be determined by using electron lines of known energy and intensity. The density of blackening, which could be obtained for the three lines, would determine the strength of the source to be used for an exposure time of a few hours.

If the 37 keV transition proved to be E2, the schemes a, c and e would be ruled out. To determine whether b or d, if either, were the correct scheme, it would be necessary to examine the multipolarity of the "169 keV" transition; scheme b would require it to be E1, whereas scheme d would require it to be a mixture of E1 (170 keV) and E2 (169 keV). If, on the other hand, the 37 keV transition were E1, indicating that

schemes a, c and e are possibilities, some information could be obtained by establishing whether or not conversion electrons were emitted in a $0^+ \longrightarrow 0^+$ transition of about 253 keV.

Chapter VI.

SEARCH FOR A RARE α-EMISSION IN ^{228}Ra (MsTh1).

VI.1. Introduction.

From time to time during the classical investigations into natural radioactivity, the possibility of a branching in the thorium series at ^{228}Ra (MsTh1) and ^{228}Ac (MsTh2) was considered. In the case of ^{228}Ac, various searches were made both for a rare α-emission and for the unknown isotope, ^{224}Fr, which would result from such a disintegration. The most careful of these, by Hahn and Erbacher (1926), led to the conclusion that the α/β branching ratio of ^{228}Ac is $< 1.5 \times 10^{-7}$, assuming that the half-life of the ^{224}Fr is less than ten years.

The results of studies of the systematics of α-disintegration in the heavy elements have been given by Perlman, Ghiorso and Seaborg (1950) and by Glass, Thompson and Seaborg (1955). From a consideration of these systematics it is possible to estimate the energies and then the half-lives to be expected in the α-disintegrations of ^{228}Ra and ^{228}Ac. By using these estimated half-lives, in conjunction with the known half-lives for the β-disintegration of ^{228}Ra and ^{228}Ac, the α/β branching ratios can be calculated (see

Table 4).

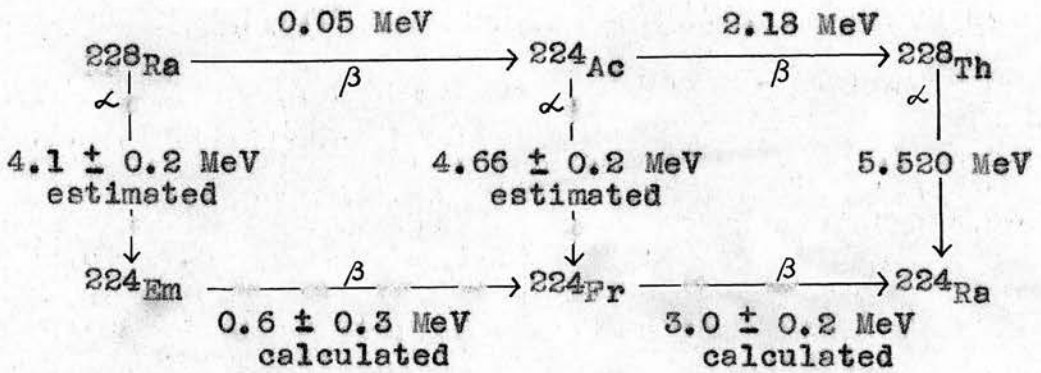
Table 4.

	Estimated α-disintegration energy	Estimated half-life for α-disintegration	Known half-life for β-disintegration	Calculated α/β branching ratio
^{228}Ra	4.10 MeV	10^9 years	6.7 years	6.7×10^{-9}
^{228}Ac	4.66 MeV	10^5 years	6.13 hours	7×10^{-9}

The α/β branching ratio of ^{228}Ra can therefore be taken as 6.7×10^{-9}, within a factor of about 150 either way due to an uncertainty of $\pm$ 0.2 MeV in the estimated disintegration energy, and that of ^{228}Ac as 7×10^{-9}, within a factor of 100 either way corresponding to a similar uncertainty. If the α-disintegration of ^{228}Ac is hindered, as it probably is, the probable α/β branching ratio is likely to be of the order of 10^{-10}. The search for the unknown products, ^{224}Em and ^{224}Fr, which result from such branching, thus presents a formidable problem. A method of great potential sensitivity is, however, available using nuclear emulsions.

By considering the closed disintegration cycles as shown below, it can be predicted that the ^{224}Em is β-unstable by 0.6 $\pm$ 0.3 MeV, and that the ^{224}Fr, which results both from the α-disintegration of ^{228}Ac and

the β -disintegration of ^{224}Em, is β -unstable by 3.0 ± 0.2 MeV.



The heavy lines indicate the principal modes of disintegration (see Figure 1) and the broken lines represent the branch disintegrations. The energies quoted are the Q-values for the various disintegrations.

In order to make accurate estimates of the partial half-lives it is necessary to know whether the β - disintegrations are allowed or forbidden. In respect of ^{224}Em, which is the more important for the present purpose, there is not much guidance to be obtained from a comparison with similar species. ^{228}Ra, with a much smaller disintegration energy of $\sim$ 0.05 MeV, disintegrates by an allowed transition, but the principal mode of disintegration of ^{234}Th (disintegration energy 0.20 MeV) is at least ten times slower than allowed. The principal mode of ^{240}U (disintegration

energy 0.36 MeV) is almost certainly allowed but that of ^{246}Pu (disintegration energy 0.40 MeV) is again some three times slower.

Some information can be obtained by making reasonable conjectures regarding the spin and parity changes which result from the β-disintegration of ^{224}Em. If it is assumed that the spin change is 0 or $\pm$ 1, then those disintegrations which involve no change of parity will be characterised by an ft-value of the order of 10^4; if the parity is changed, then the relevant ft.-value will be either $10^{5.5}$ or $10^{6.5}$, corresponding to a spin change of 0 or $\pm$ 1 respectively. Such spin changes are probable since the ground state of ^{224}Em, from which the β-disintegration occurs, has a spectroscopic designation of 0 even, and it is likely that either the ground state of ^{224}Fr or an excited level to which the β-disintegration could lead on the basis of the available energy, will have a spin of 0 or 1. If the principal disintegration mode of ^{224}Em is to the ground state of ^{224}Fr, then the half-life could be as short as a few minutes (no parity change) or as long as two days (spin change of 1 and a parity change); on the other hand, if the principal mode of disintegration has an energy of, say, 0.4 MeV the half-life could be as long as six days.

In respect of ^{224}Fr, a close estimate is not

necessary for the present purpose. Since, however, 3.0 MeV is available for the disintegration and since there are, at least, four energy levels close to the ground state in ^{224}Ra, the half-life is probably a few minutes only, although it could be as long as a few hours. Therefore, whatever the half-lives, within the above-stated limits, the α-branching at ^{228}Ra will result in a fairly rapid return to the main disintegration chain at ^{224}Ra.

The fact that the ^{224}Em is also α-unstable is not worth taking into consideration in the present investigation, since the predicted half-life is of the order of 100 years.

VI.2. Experimental Method.

The method makes use of the fact that all the isotopes of the emanation, which have grown within a solution of ^{228}Ra in a given time, can be isolated in a liquid air trap. Any ^{224}Ra, which then grows in this isolated fraction, can have been produced only by the disintegration of the ^{224}Em. By impregnating a nuclear emulsion with this residue the presence of ^{224}Ra can be detected with certainty by virtue of the fact that its disintegration results in the formation of a "star" consisting of three or four α-particle tracks (see Figure 1). Any background of active

deposit, from the disintegration of ^{220}Em, could not give rise to such "stars". The bulk of the unwanted activity was removed, by simple chemical separations, before impregnation.

In the present experiments, a solution containing 2 mC of ^{228}Ra in 5 ml of 1N hydrochloric acid, was allowed to stand in a sealed system for at least fourteen days, to allow any ^{224}Em present to approach secular equilibrium even on the longest estimate of its half-life. Nitrogen was then bubbled through the solution for four hours. The emerging gas stream passed up a column, 2 cm. in diameter, filled with glass beads and glass wool, and then through a trap, containing 2 ml of a frozen 1N nitric acid solution, surrounded with liquid oxygen. The efficiency of the column in trapping spray had been checked previously by bubbling nitrogen through a solution containing 5 mC of ^{212}Pb and examining the condensate for this activity with nuclear emulsion; no such activity was found. In the experiments with ^{228}Ra, the trap was sealed after the bubbling had been stopped, allowed to stand for twenty-four hours, during which time it warmed up and the residue dissolved in the thawing nitric acid solution.

A few drops of this solution was removed and set aside for fourteen days to allow the ^{212}Pb to

disintegrate. It was then used to impregnate a nuclear emulsion as an additional check on any carry-over of the parent activity, in spray, or by volatilisation. The fact that no such activity, which would be indicated by the presence of "stars" consisting of five α-particle tracks, was observed proved that the carry-over was negligible. The ^{222}Em, which results from the disintegration of ^{226}Ra present in the ^{228}Ra, was removed by evaporating the main solution to dryness. In any case, the disintegration products of ^{222}Em could not confuse the issue by giving rise to "four-track stars". The residue was then redissolved in nitric acid, carriers of barium and caesium were added and were separately freed from traces of lead, bismuth and thorium isotopes; in the former case by eight successive precipitations of the barium as nitrate by fuming nitric acid in the presence of lead and bismuth carriers; in the latter by eight successive precipitations as carbonates of barium, lead and bismuth carriers added to the solution, followed by the precipitation of caesium as perchlorate.

The purified solutions of barium and caesium were used to impregnate Ilford G5 emulsions and these were left to stand for fourteen days. If the half-life of the intermediate ^{224}Fr were short, then the emulsion impregnated with the barium solution would contain

initially the bulk of the ^{224}Ra which results from the branching. If, however, the half-life of the ^{224}Fr were, unexpectedly, greater than two days, then the ^{224}Ra would accumulate in the emulsion impregnated with the caesium solution. Emulsions were impregnated with the distilled water used in the separation processes and with solutions of the barium and caesium carriers; these emulsions were used as controls.

Freshly prepared emulsions were used in this investigation in order that the number of background events, inherent in the emulsion, would be as small as possible. The final volume of the solutions, used to impregnate the emulsions, was 25 cc.; the reason for the use of such a large volume was the fact that the emulsion could not absorb a concentration of barium greater than 1 mg. per 5 cc. The emulsions were impregnated, exposed and processed in the same manner as described in Chapter II. Searching of the plates was performed under a low-power air objective and measurements, on some of the events found, were made under a high power oil-immersion objective. In each plate an area of 2 cm.^{2} was examined. Since the emulsion swells to about ~~twice~~ three times its normal thickness on impregnation, this area corresponded to a volume of about 8×10^{-2} cc.

VI.3. Results and Conclusions.

Four runs of this type were made and each time a weak activity was detected in the emulsions which had been impregnated with Ba + ^{224}Ra solution. This activity was identified by a significant increase in the "stars" consisting of four α-particle tracks unaccompanied by similar increases in the numbers of "stars" with three or five α-particle tracks (see Table 5).

Table 5.

	3-track "stars"	4-track "stars"	5-track "stars"
Ba + Ra plates			
1st run	6	15	5
2nd run	15	28	10
3rd run	9	30	8
4th run	7	18	6
Totals	37	91	29
Barium plates			
1st run	4	7	4
2nd run	9	12	9
3rd run	13	18	16
4th run	6	11	4
Totals	32	48	33
Differences (with standard deviations)	5 ± 8	43 ± 12	(-) 4 ± 8

Since the difference of 43 is more than three times its standard deviation it was considered to be significant. Furthermore, since about two-thirds of these "stars" showed an α-particle track with a range of 47μ, corresponding to the 8.78 MeV disintegration of ^{212}Po, they must have resulted from the successive disintegrations starting at ^{224}Ra (see Figure 1). No activity, significantly higher than the background, was found in the emulsions which had been impregnated with the Cs + ^{224}Fr sclution and this suggested that the half-life of the ^{224}Fr is less than twenty-four hours.

Until the intermediate isotopes ^{224}Em and ^{224}Fr are identified, these results cannot be considered as proof that α/β branching occurs at ^{228}Ra, but they do allow the placing of upper limits on the value of the branching ratio. These upper limits were calculated by making various assumptions regarding the half-lives of the ^{224}Em and the ^{224}Fr. The half-life of ^{224}Fr, which is less than twenty-four hours, may be as short as a few minutes or as long as about twelve hours and still give rise to no significant increase in the emulsions impregnated with the Cs + Fr solution. The values quoted in Table 6 take account of the fact that there were two sources of ^{224}Em in the condensates (i) that which had accumulated in the fourteen day period of standing before the bubbling started and (ii) that

which had accumulated in the four hour bubbling period. They also include a factor of five to cover possible losses in the condensation and separation processes and a factor of three hundred to take account of the fact that only a small fraction of the impregnating solution was absorbed by the emulsion.

Table 6.

Upper limits of the α/β branching ratio of ^{228}Ra.

Estimated half-life of ^{224}Em	Estimated half-life of ^{224}Fr:	Upper limit 5 minutes	Upper limit 12 hours
6 minutes		2×10^{-8}	3×10^{-8}
1 hour		1.5×10^{-8}	2×10^{-8}
10 hours		5×10^{-9}	7×10^{-9}
100 hours		3×10^{-9}	4×10^{-9}
200 hours		3×10^{-9}	3×10^{-9}

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